



Project Number 282910

## ÉCLAIRE

## Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems

Seventh Framework Programme

Theme: Environment

## **Project Final Report**

Due date of deliverable:01/12/2015

Actual submission date: 01/12/2015

Start Date of Project: 01/10/2011

Duration: 48 months

Organisation name of lead contractor for this deliverable:

### Natural Environment Research Council

Pr	Project co-funded by the European Commission within the Seventh Framework						
	Dissemination Level						
PU	Public	х					
PP	Restricted to other programme participants (including the Commission						
RE	Restricted to a group specified by the consortium (including the						
со	Confidential, only for members of the consortium (including the Commission Services)						

### Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems

## **Final Report**

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1 December 2015

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## Part 1: Final Publishable Summary Report

## **1. Executive Summary**

The central goal of ECLAIRE is to assess how climate change will alter the extent to which air pollutants threaten terrestrial ecosystems. Particular attention has been given to nitrogen compounds, especially nitrogen oxides ( $NO_x$ ) and ammonia ( $NH_3$ ), as well as Biogenic Volatile Organic Compounds (BVOCs) in relation to tropospheric ozone ( $O_3$ ) formation, including their interactions with aerosol components. ECLAIRE has combined a broad program of field and laboratory experimentation and modelling of pollution fluxes and ecosystem impacts, advancing both mechanistic understanding and providing support to European policy makers.

## The central finding of ECLAIRE is that future climate change is expected to worsen the threat of air pollutants on Europe's ecosystems.

Firstly, climate warming is expected to increase the emissions of many trace gases, such as agricultural  $NH_3$ , the soil component of  $NO_x$  emissions and key BVOCs. Experimental data and numerical models show how these effects will tend to increase atmospheric N deposition in future. By contrast, the net effect on tropospheric  $O_3$  is less clear. This is because parallel increases in atmospheric  $CO_2$  concentrations will offset the temperature-driven increase for some BVOCs, such as isoprene. By contrast, there is currently insufficient evidence to be confident that  $CO_2$  will offset anticipated climate increases in monoterpene emissions.

Secondly, climate warming is found to be likely to increase the vulnerability of ecosystems towards air pollutant exposure or atmospheric deposition. Such effects may occur as a consequence of combined perturbation, as well as through specific interactions, such as between drought, O<sub>3</sub>, N and aerosol exposure.

These combined effects of climate change are expected to offset part of the benefit of current emissions control policies. Unless decisive mitigation actions are taken, it is anticipated that ongoing climate warming will increase agricultural and other biogenic emissions, posing a challenge for national emissions ceilings and air quality objectives related to nitrogen and ozone pollution. The  $O_3$  effects will be further worsened if progress is not made to curb increases in methane (CH<sub>4</sub>) emissions in the northern hemisphere.

Other key findings of ECLAIRE are that: 1) N deposition and  $O_3$  have adverse synergistic effects. Exposure to ambient  $O_3$  concentrations was shown to reduce the Nitrogen Use Efficiency of plants, both decreasing agricultural production and posing an increased risk of other forms of nitrogen pollution, such as nitrate leaching (NO<sub>3</sub><sup>-</sup>) and the greenhouse gas nitrous oxide (N<sub>2</sub>O); 2) within-canopy dynamics for volatile aerosol can increase dry deposition and shorten atmospheric lifetimes; 3) ambient aerosol levels reduce the ability of plants to conserve water under drought conditions; 4) low-resolution mapping studies tend to underestimate the extent of local critical loads exceedance; 5) new dose-response functions can be used to improve the assessment of costs, including estimation of the value of damage due to air pollution effects on ecosystems, 6) scenarios can be constructed that combine technical mitigation measures with dietary change options (reducing livestock products in food down to recommended levels for health criteria), with the balance between the two strategies being a matter for future societal discussion. ECLAIRE has supported the revision process for the National Emissions Ceilings Directive and will continue to deliver scientific underpinning into the future for the UNECE Convention on Long-range Transboundary Air Pollution.

## 2. Summary of Project Context and Objectives

Exceedances of threshold levels for ecosystem impacts of ozone (O<sub>3</sub>) result in significant impacts on semi-natural ecosystems, while an estimated ~ $\in$ 7 billion in the year 2000 were lost due to phyto-toxic impacts of O<sub>3</sub> on arable crops (Holland et al., 2006). Due to intercontinental transport, future O<sub>3</sub> concentrations will depend crucially on how emissions of nitrogen oxides (NO<sub>x</sub>) and volatile organic compounds (VOCs) evolve in the developing world, outside Europe, but it is likely to have severe implications for the economy and global food security (Derwent et al., 2004; Ashmore et al., 2005; Royal Society, 2008).

At the same time, atmospheric reactive nitrogen compounds (N<sub>r</sub>) represent an increasingly important pollution driver of European land ecosystems, especially as emissions of sulphur dioxide (SO<sub>2</sub>) in the EU-27 decreased by nearly 70% between 1990 and 2007, with much smaller reductions for NO<sub>x</sub> (~30%) and ammonia (NH<sub>3</sub>, ~15%) over the same period. With latest data reported to 2013, the reductions since 1990 are 87% for SO<sub>2</sub>, 55% for NO<sub>x</sub> and 28% for NH<sub>3</sub> (CEIP, 2015).Together, wet and dry deposition of both oxidized and reduced nitrogen are having multiple impacts on terrestrial ecosystems, in some cases increasing productivity and carbon storage (de Vries et al., 2009). However, nitrogen deposition also is threatening ecosystem functioning and plant community composition in many areas (Bobbink et al., 2010), representing an annual loss of ~€10-70 billion (TFRN, 2010a; Brink et al. 2011).

Last but not least, many atmospheric pollutants that affect ecosystems, like ozone, nitrogen and secondary aerosols, are not only important climate forcing agents (Andreae et al., 2005; Arneth et al., 2009; Forster et al., 2007), but their atmospheric burden strongly responds to climate change in turn (Dentener et al., 2006b; Johnson et al., 2001; Racherla & Adams, 2006). The interactions of climate change, change in nitrogen deposition, increasing atmospheric CO<sub>2</sub> concentration, changing aerosol burdens and changing ozone background and peak levels make projections of pollution impacts on terrestrial ecosystems challenging. This is especially so, since these affect ecosystem physical and biogeochemical responses on different spatial and temporal scales, and individually in either positive or negative ways (e.g., on ecosystem productivity, water use efficiency, carbon storage or biodiversity; Arneth et al., 2010a; Mercado et al., 2009; Sitch et al., 2007; Pleijel et al., 2014, Simpson et al., 2014a). What is more, changing biogenic emissions in response to air pollution and/or climate change can affect air pollution and climate change in turn, in a complex system that contains multiple, interacting feedbacks (Arneth et al., 2010b; Raes et al., 2010).

To optimise the efficacy of European emission control strategies in the global pollution-climate change context, it is vitally important that we develop a consistent and process-based observational and modelling framework to understand how interactive atmospheric pollutants will impact ecosystems in response to climate and air pollution change.

Focusing especially on the role of ozone and nitrogen, and where relevant their interactions with volatile organic compounds, aerosols and sulphur, the Overall Objectives of ECLAIRE are therefore:

- O 1. to provide robust understanding of air pollution impacts on European land ecosystems including soils under changing climate conditions, and
- O 2. to provide reliable and innovative risk assessment methodologies for these ecosystem impacts of air pollution, including the economic implications, to support EU policy.

ECLAIRE targets climate-ecosystem-atmosphere interactions and their implications for ecosystem effects at the European scale, combining observations and experiments in the field and laboratory with modelling experiments from plot to European scales, while accounting for changes in global background.

Such new scientific understanding and risk-assessment methodologies under climate change is of central importance in the current EU negotiations under the Convention on Long-range Transboundary Air Pollution (CLRTAP). Already, with recent revision of the Gothenburg Protocol, air pollution - climate interactions have begun to be addressed prior to ECLAIRE (e.g., TFRN, 2010b; Sutton et al., 2011). Given the need to quantify the policy co-benefits, the outputs of ECLAIRE were envisaged as being even more important in supporting the CLRTAP 'Long-Term Strategy' (UNECE, 2010a).

To reach its Overall Objectives, ECLAIRE has addressed the following Key Questions:

- Q1: What are the expected impacts on ecosystems due to changing ozone and N-deposition under a range of climate change scenarios, taking into consideration the associated changes in atmospheric CO<sub>2</sub>, aerosol and acidification?
- Q2: Which of these effects off-set and which aggravate each other, and how do the mitigation and adaptation measures recommended under climate change relate to those currently being recommended to meet air pollution effects targets?
- Q3: What are the relative effects of long-range global and continental atmospheric transport vs. regional and local transport on ecosystems in a changing climate?



Fig. 1. Schematic of ECLAIRE highlighting the main science Components and Work Packages.

- Q4: What are the appropriate metrics to assess ozone and nitrogen impacts on plants and soils, when considering state-of-the-art understanding of interactions with CO<sub>2</sub> and climate, and the different effects of wet vs. dry deposition on physiological responses?
- Q5: What is the relative contribution of climate dependence in biogenic emissions and deposition vs. climate dependence of ecosystem thresholds and responses in determining the overall effect of climate change on air pollution impacts?
- Q6: Which mitigation and/or adaptation measures are required to reduce the damage to "acceptable" levels to protect carbon stocks and ecosystem functioning? How do the costs associated with the emission abatement compare with the economic benefits of reduced damage?
- Q7: How can effective and cost-efficient policies on emission abatement be devised in the future?

To be able to answer these questions the project focuses on improving the understanding of the interactions and feedbacks in the coupled biosphere-chemistry-climate system and developing novel approaches to quantifying ecosystem effects and threats together with improved tools for upscaling to Europe and extrapolating to future climates. The integration of these issues has focused on the following **Specific Objectives** (for Work Package numbers see **Figure 1** – 'Month', refers to the completion month for work concerning each Specific Objective):

- S1. To develop improved process-based emissions parameterization of NH<sub>3</sub>, NO and VOCs from natural and agricultural ecosystems in response to climate and pollutant deposition for incorporation into atmospheric Chemistry-Transport Models (CTMs), based on existing and new flux measurements in the field and laboratory, applying these to develop spatially resolved emission scenarios in response to climate, CO<sub>2</sub> and air pollutant change [WPs 1, 2, 3, 6./Month 42].
- S2. To determine the chief processes in atmospheric chemistry that respond to climate and air pollution change and the consequences for ozone and aerosol production and atmospheric lifetimes, in the context of the global O<sub>3</sub> background [WPs 5, 7/Month 36 & through collaboration with PEGASOS FP7 project].
- S3. To develop improved multi-layer dry deposition / bi-directional exchange parameterisations for O<sub>3</sub>, NO<sub>x</sub>, NH<sub>3</sub>, VOCs and aerosols, taking into account near-surface chemical interactions and the role of local/regional spatial interactions, based on existing and new flux measurements and high resolution models and to estimate European patterns of air concentrations and deposition under climate change [WPs 1, 2, 4, 7, 8/Month 42].
- S4. To integrate the results of meta-analyses of existing datasets with the results of targeted experiments for contrasting European climates and ecosystems, thereby assessing the climate-dependence of thresholds for land ecosystem responses to air pollution, including the roles of ozone, N-deposition and interactions with VOCs, nitrogen form (wet/dry deposition) and aerosol [WPs 9, 10, 11, 12/Month 30].
- S5. To develop improved process-based parameterizations in dynamic global vegetation models (DGVMs) and soil vegetation models (DSVMs) to assess the combined interacting impacts of air quality, climate change and nutrient availability on plant productivity, carbon sequestration and plant species diversity and their uncertainties [WP13; WP14; WP15, WP17/Month 44].
- S6. To develop novel thresholds and dose-response relationships for air pollutants (especially for O<sub>3</sub> and N) under climate change, integrated into process-based models verified by

experimental studies at site scales and mapped at the European scale, quantifying the effect of climate change scenarios [WPs 12, 13, 14, 15, 16/Month 44].

- S7. To assess the extent to which climate change alters the transport distance and spatial structure of air pollution impacts on land ecosystems considering local, regional, continental and global interactions, focusing on nitrogen and ozone effects [WPs 5, 6, 7, 8, 9/Month 44].
- S8. To apply the novel metrics to quantify multi-stress response of vegetation and soils, including effects on carbon storage and biodiversity to improve the overall risk assessments of pollution-climate effects on ecosystems at the European scale as the basis for development of mitigation options [WPs 12, 13, 14, 15, 16, 19, 20/Month 44].
- S9. To quantify the overall economic impacts of air pollution effects on land ecosystems and soils, including the valuation of ecosystem and other services, and the extent to which climate change contributes by altering emissions versus ecosystem vulnerability [WPs 3, 4, 6, 7, 12, 14, 15, 16, 18/Month 42].
- S10. To reassess the current recommendations regarding air pollution emission abatement policies, considering the interactions between ecosystem and other effects under conditions of climate change and to perform cost-benefit analysis of policy options under different scenarios [WPs 18, 19, 20/Month 48].

As can be seen from Figure 1, ECLAIRE is organised around **five chief science components**, supported by a smaller number of strategic and management actions, to provide end-to-end science from measurements and improved process understanding, over model integration, to the advice on mitigation and adaptation strategies. The work packages in each component are designed to provide novel understanding from small-scale biogeochemical processes to European and global simulations.

Component 1 derives the process understanding to link biogenic/agricultural emissions and deposition to vegetation and soils, to meteorological conditions and to pollutant inputs, through meta-analysis of existing flux data, fluxes from a 9-site flux network across the European climate space and targeted controlled measurements of emission, deposition and chemical conversion processes. The emerging parameterisations are used in **Component 2** to develop improved, more mechanistic, modelling frameworks that simulate the effect of the interactions of the climate-atmosphere-biosphere system on biogenic emission and bi-directional exchange, providing emission, deposition and concentration fields at the European scale that respond to global change. Using these exposure and deposition maps, and data from ecosystem manipulation experiments addressing air pollution - climate interactions, **Component 3** has worked to improve dose/response relationships under changing climate, develop new thresholds and improved models to simulate the effect of pollutants on aboveand below-ground carbon stocks. Upscaling of ecological responses, thresholds and exceedances to the regional and European scale and its spatial variability is provided by Component 4, while the implications for the economy and ecosystem services is assessed by Component 5, which also considers the implications for mitigation and adaptation strategies.

## 3. Main Science and Technology Results

### 3.1. Component 1: Emissions & Exchange Processes

Component 1 has improved understanding of the exchange of pollutants that are directly or indirectly relevant for ecosystem effects between the atmosphere and the vegetation through measurements across a European ten-site flux network (nine funded and one providing data as an associated site) and target laboratory studies. It then used the new data, together with existing datasets from previous European and national projects, to develop improved models and parameterisations of the exchange processes for use in spatial chemistry and transport as well as chemistry and climate models that are used to map the deposition, exposure and impacts of air pollutants. Here, rather than improving static parameterisations and parameter look-up tables that have been compiled for current conditions, the focus of ECLAIRE has been to develop models that can increasingly capture the response to changes in climate and atmospheric composition and account of pollutant interactions.



**Fig. 2:** Overview of the 10 measurement sites of the European ECLAIRE flux network that were operated for 15 months covering 2013. The blue dots indicate the location of additional short-term campaigns that were fully or partly supported by ECLAIRE.

### Field flux measurements and interpretation

The 10-site network (**Figure 2**) aimed to measure fluxes of ozone and nitrogen oxides continuously for a duration of 15 months, including the calendar year 2013. This provided the first synchronised multi-site ozone flux dataset to date, measured with a harmonised (eddy-covariance) approach. The ozone flux data have been used (a) to assess how much of the  $O_3$  enters the plant stomata where it can cause damage, (b) to improve three  $O_3$  deposition models with different emphases and (c) to look for **field evidence of the immediate effect of**  $O_3$  **on plant growth**. Castelporziano in Italy, where  $O_3$  effect of growth was significant throughout

the year. At Speulder forest it was significant in summer. However, if average concentration over the previous 24 hours was used as a driver, the effect became significant at all sites.

In addition to the long-term measurements, fluxes of volatile organic compounds (VOCs, as important precursors for ozone and formation of particulate matter), particles and ammonia were measured for shorter periods at selected sites of the site network and beyond (**Figure 2**). These measurements have revealed that **moorland vegetation can be a large source of isoprene on warm days and would gain in importance with global warming**. They have also greatly enhanced the emission factor database used to calculate isoprene emissions from oak trees, which are thought to account for 60% of Europe's plant isoprene emissions.

As part of the flux network, a new major forest tower was established at the site of Bosco Fontana (BF), a hornbeam-oak woodland in the NE Po Valley, Italy, which hosted a collaborative flux campaign, which was co-ordinated with concentration measurements elsewhere in the Po Valley made by the FP7 PEGASOS as well as the Italian SUPERSITO project. This BF campaign was designed to bring a large amount of instrumentation from different institutes together to quantify the deposition and emission of pollutants in this polluted part of Europe and to study the importance of chemical interactions between pollutants within and above plant canopies (in this case a forest) for changing the deposition loads experienced by the vegetation (Acton et al., 2015; Schallhart et al., 2015). See **Figure 3**.



**Fig. 3:** Schematic of the instrument setup during the ECLAIRE integrated campaign at Bosco Fontana.

The data indicate that the atmospheric nitrogen load to this site is very high, with an extrapolated annual figure of nearly 75 kg N ha<sup>-1</sup> yr<sup>-1</sup>, dominated by ammonia (NH<sub>3</sub>), plus an additional contribution through precipitation (Twigg et al., 2015). The state-of-the-art measurements have provided additional evidence that some nitrogen-containing components found in particulate matter dissociate into gases during the deposition process. Because gases deposit much faster than the particles this process

exacerbates total N deposition (with adverse effects on biodiversity) and reduces aerosol concentrations (with positive impacts on reducing human health effects). Together with similar measurements from other sites (Speuld, Auchencorth), a first empirical parameterisation was developed for inclusion into chemistry and transport models, which indicates that the effect lowers fine nitrate concentrations significantly (by 30% at the European and annual average) (Nemitz et al., 2014).

The high N load at BF results in very high soil emissions of nitric oxide (NO), the reaction with which accounts for 30% of the total ozone sink of the canopy (see **Figure 4**, Finco et al. 2015). Whilst this has the potential of mitigating the impact of ozone on the forest, much of this appears to occur in the understorey, thus leaving the  $O_3$  burden to the tree canopy unmodified.





### Controlled environment studies and model development

Lab-based gas-exchange measurements using soil cores and leaf litter from Bosco Fontana and the other sites of the network developed new relationships between emissions of NO and  $NH_3$  (as well as the greenhouse gases  $CO_2$  and  $CH_4$ ) and meteorological drivers (soil moisture and temperature). The investigations revealed that **ground NO emissions at BF and some other sites are dominated by the litter rather than the soil**, and this has important consequences for the future improvement of process-based models that often do not explicitly treat the litter layer.

ECLAIRE has also identified that **plants produce NO internally in response to environmental stresses.** The likely function of this NO is to communicate the presence of stress across the plant. However, levels are too low to be atmospheric relevance.

To improve the prediction of NO emissions from (mineral) soils ECLAIRE has completely rewritten the DNDC model into 'Landscape DNDC' to prepare it for application in a spatial context, and replaced its soil biogeochemistry module, whose parameter settings were tuned and then validated against a large combined flux measurements of NO and N<sub>2</sub>O from a range of projects including ECLAIRE.

Other lab-based measurements, using a unique coupling of a plant chamber and a smog chamber for chemistry experiments, have sought to take a holistic view of the net effect of trees on ozone. Whilst  $O_3$  is removed by deposition to plants locally, the VOCs emitted by plants have the potential to result in  $O_3$  formation downwind. The measurements have shown that the net effect greatly differs between tree species and is highly sensitive to the NO<sub>x</sub> regime (i.e. remoteness of the site from industrial and traffic sources).

The same setup was used to study the effect of biotic stress (insect attack) on the quantities and chemical makeup of VOCs released by plants. Aphid attack was found to trigger the emission of VOCs that are particularly effective in producing particulate matter and this effect was scaled up to Europe as an exploratory exercise (Mentel et al., 2013; Bergström et al., 2014). These first investigations into the effect suggests that the effect of climate change on the frequency of biotic stresses may affect future VOC emissions and their impacts.

Lab investigations also produced new functional dependencies of emissions of monoterpene (the second most important class of VOCs after isoprene) on drought stress and investigated the controls of the exchange of isoprene oxidation products with plants.

Stomatal conductance, which is regulated by the plant's need to take up carbon whilst controlling water loss, influences the emission and deposition of many gases and in particular controls the amount of ozone that can enter the leaf where it can cause damage and reduce plant growth. Thus, for the improvement of ozone exchange modelling, one particular focus was to incorporate into the biosphere / atmosphere exchange models that underpin European CTMs a more mechanistic stomatal conductance model. The existing DOS3E model was updated with a stomatal conductance model that is coupled to photosynthesis model and this allowed the impacts of leaf nitrogen status and ozone dose to be incorporated. This was a vital step for preparing the model for the O<sub>3</sub> and N impact assessments of Components 3 and 4. ECLAIRE has also made progress in understanding (and parameterising) the control of the non-stomatal ozone sinks, i.e. deposition to soils and leaf-surface reaction with antioxidants leached from leaves during senescence (Poitier et al. 2015).

To provide an improved, climate-sensitive representation of ammonia emission from fertilisation events and its exchange with agricultural and semi-natural vegetation more generally, an existing parameterisation was tested and refined further against a large number of NH<sub>3</sub> measurement datasets. Then two meta-models were developed as an approximation to a large number of runs conducted with a detailed ammonia volatilisation model (Volt'Air) and a crop model (CERES-EGC), to predict the fractional amount and timing of fertiliser emission and the ammonia emission potentials from soil and foliage in response to N inputs, respectively. The resulting model is based on detailed process modelling, whilst being sufficiently computationally efficient for incorporation into regional scale CTMs and capturing the key dynamics expected under climate change.

Going beyond the original work plan, ECLAIRE embarked on the ambitious activity of developing an ECLAIRE Ecosystem Surface eXchange (ESX) model as a coupled multilayer exchange, chemistry and transport model (Simpson and Tuovinen, 2014, 2015). ESX is designed to be run stand-alone at site level and also be coupled to the European EMEP CTM and its modular design allows it to be used with state-of-the-art parameterisations against which simplified expressions can be compared and optimised. This coupled model can explicitly simulate chemical interactions and storage of pollutants within the canopy air space, e.g. to simulate the chemical interactions observed during the BF campaign, and has the potential to provide a true step-change in the description of atmosphere/biosphere interactions. Due to its complexity it requires further comprehensive measurement datasets of the type collected at BF to parameterise and constrain it more fully and it will be further developed beyond ECLAIRE.

### Measurement methods and data treatment

As part of the work under Component 1, ECLAIRE has made important contribution to the development of measurement approaches and instrumentation that will have without doubt an important legacy beyond the project lifetime: ECLAIRE has developed new lab facilities and the forest flux tower at BF. It has, for the first time, standardised field flux measurements of ozone and to some extent VOCs and improved the quality control procedures associated with such measurements. It has contributed to the improvement to the first eddy-covariance flux measurements for aerosol chemical components and discovered a new (VOC) interference in a type of commercial O<sub>3</sub> analyser. ECLAIRE has also improved the retrieval mechanisms and error calculations to derive atmospheric column NH<sub>3</sub> concentrations from satellite observations (**Figure 5**). Validation is difficult between measurements and retrievals derive different entities that can only be linked via a robust CTM (van Damme et al., 2015).



**Fig. 5:** Comparison of satellite retrievals of total column NH<sub>3</sub> concentration ( $10^{16}$  molec cm<sup>-2</sup>; right colour scale) with surface concentrations measured under NitroEurope IP (NEU,  $\mu$ g m<sup>-3</sup>; left colour scale). Left panel: annual data in the spatial context; Right panel: correlation of monthly values.

# 3.2. Component 2: Emissions & Exchange at Local, European to Global Scales

Component 2 aims to: (1) provide past-to-future simulations of European to global-scale level pollution-climate change interactions, accounting for local and long-distant pollution source contributions; (2) assess how biogenic pollutants and precursors from natural, semi-natural and agricultural ecosystems vary in space and time; (3) apply the analyses of climate change-pollution interplay to combine novel knowledge into pollution metrics across Europe; (4) investigate climate-pollution interplay at high spatial resolution to take into consideration effects of landscape heterogeneity. The main type of models used in C2 were dynamic global vegetation models (LPJ-GUESS, ORCHIDEE) and chemical transport models (CTMs: LMDZ-INCA, EMEP MSCW, MATCH, DEHM etc., see Langner et al., 2012; Simpson et al, 2014b; Schaap et al., 2015). We summarise below the main results.

### Emissions and ecosystems in a changing climate

Agriculture is clearly a large source of various N-containing trace gases, especially in response to fertilisation, and a process of newly recognized significance is that **a warmer climate will increase NH**<sub>3</sub> **emissions from sources such as animal manure**. In order to deal with such effects, a new ammonia model has been developed that provides dynamic emissions in response to climate change (warmer temperatures enhancing NH<sub>3</sub> emissions notably) and fertilisation (Sutton et al., 2013; Skjöth & Geels, 2013; Werner et al., 2015). Work has also been done in order to implement the new and improved dynamical NH<sub>3</sub> emission model in some of the Chemistry-Transport models (CTMs) used in ECLAIRE. These studies suggest that the effect of weather and climate change on the emissions of ammonia is currently underestimated in existing ammonia emission models.

The impact of climate on biogenic volatile organic compound (BVOC) emissions is more complex (Messina et al., 2015; Arneth et al., 2010; Simpson et al., 2014a). There is a clear finding **that increasing temperatures drives increases in emissions** (see also **Figure 18**). However, **increasing CO**<sub>2</sub> **may cause decreases for some BVOCs, especially isoprene** which has a high ozone-forming potential. By contrast, there is much less certainty on whether CO<sub>2</sub> will offset the warming effect for monoterpenes. Globally, comparing the 2040s and the 1990s, ORCHIDEE calculations indicate 25% increases in isoprene, 27% in monoterpenes and 28% in methanol emissions. However, inclusion of the CO<sub>2</sub> effect completely off-sets the increase in isoprene emissions for the RCP4.5 GHG scenario, indicate isoprene emissions increase 41% and monoterpenes 25% in the future compared to current conditions. However, taking the CO<sub>2</sub> inhibition effect into account, emissions decrease slightly with -2% and -13% respectively for isoprene and monoterpenes in LPJ-GUESS. As well as climate factors, BVOC emissions are affected by land-cover changes.

LPJ-GUESS calculations of wildfire emissions indicate a **complex range of interactions between vegetation, climate change and increasing CO<sub>2</sub>, and fire suppression**. Comparing 1970-2000 and 2070-2100, overall tropical emissions decline between 15 and 35 % (mostly due human influence), while extratropical emissions increase by 20 % and 45 %. Globally emissions change within a -10 % range.

While nitrogen input to ecosystems affects yields and can lead to pollution of watersheds in heavily fertilised regions, the DGV models suggest only small effects of N deposition on the historical carbon sink strength of natural ecosystems. Whether or not nitrogen limitation of plant growth will notably affect future ecosystem carbon storage is under debate, and current modelling studies show conflicting results. Arguably, climate effects of N<sub>2</sub>O emissions are of more concern than N-interactions with the C sink; this will be investigated further in the coming years with updated versions of LPJ-GUESS.

LPJ-GUESS was updated with a coupled carbon-nitrogen cycle in the crop module of the code. This allows to assess impacts of N fertiliser on a range of ecosystem processes, and ecosystem services derived from these such as yields, carbon storage of nitrogen leaching. Recently simulation experiments were performed to study how different crop management options would affect the calculated values for these three services in comparison with a standard simulation set-up. Trade-off analyses of these indicated that –regionally and globally-**no-tillage would have relatively small effects on soil C pool size**, contributing to recent debate in the literature. Such results are preliminary with development continuing.

### **Atmospheric Modelling Results**

Impacts of air pollution on European ecosystems occur over a range of spatial scales from the global scale ( $O_3$  background), though regional scale ( $O_3$  and N deposition) to local scale (N deposition and  $PM_{2.5}$ ,  $NH_3$  exposure). In a changing climate, the spatial patterns of impacts are likely to change due to changing emissions, land use and atmospheric processes. Modelling carried out by ECLAIRE in cooperation with other projects (e.g. CLRTAP Task Force Hemispheric Transport of Air Pollution, TF HTAP) allows us to address these interactions.

Model results show that **90-95% of impacts due to N deposition to European ecosystems are the result of European emissions**. At a national level N deposition has contributions from both national emissions as well as emissions from neighbouring countries.

The trends of inflow of ozone at Europe's boundary is only partly understood. Possible explanations include important roles of decadal scale variability, and possibly missing information on long-range transport of anthropogenic pollution. However, it is well established that the impacts of O<sub>3</sub> in Europe are the result of precursor emissions both from within Europe and worldwide. Summertime ozone concentrations in Europe are strongly influenced by European precursor emissions whereas non-European precursor emissions, of which methane is key, dominate the rest of the year. Indeed, controlling methane and air pollution emissions in Asia is going to be of critical importance for ozone air quality in Europe.

CNRS has performed various sets of future simulations with the LMDz-INCA model (with BVOC emissions from ORCHIDEE) that have been used to investigate the future direct radiative forcing of nitrate particles (Hauglustaine et al., 2014; Messina et al., 2015). Figure 6 shows the change in surface ozone in summer associated with future emissions calculated with climate change and land use effect with or without  $CO_2$  inhibition. This figure shows that including the  $CO_2$  effect on isoprene emission responses significantly reduces the extent of ozone increase in the future.

Selected scenarios of future emissions provided by IPCC AR5 RCPs were used to evaluate the possible global, hemispheric and European evolution of ozone and other air pollutants for 2030, 2050 and 2100. Model calculations using the RCP scenarios suggest that the aerosol sulphate ( $SO_4^{2-}$ ) burden will decline strongly, while nitrate ( $NO_3^{-}$ ) and ammonium ( $NH_4^+$ ) aerosol burdens are more constant. Agricultural emission of  $NH_3$  may therefore maintain higher levels of cooling than assumed in previous studies. These results are driven by increasing agricultural  $NH_3$  emissions as defined in the RCP emission scenario does not include the climate effect on  $NH_3$  emissions identified by ECLAIRE (see above). Including the temperature effect will further emphasize the contribution of  $NH_3$  emissions to PM formation in future.

Comparison of the summer ozone distributions between 2050 and 2010 using the ECLIPSE5.0 emission scenario indicates ozone decreases by up to 7 ppbv in Northern America and by 4-5 ppb in Europe. Climate and land use change by 2050 may augment isoprene emission and lead to ozone increases in large portions of the Northern Hemisphere up to 4.5 ppb, potentially off-setting the ozone reductions by anthropogenic emissions in Europe and North America. However, when including the effect of increasing  $CO_2$  on reducing the isoprene emissions, the effect on ozone is much less, with two current parameterizations strongly disagreeing.



**Fig. 6:** Changes in ozone surface concentrations 2050 from present-day for June-July-August (ppbv). Top: climate change and land-use changes considered. Bottom: climate change, land use and  $CO_2$  inhibition considered. Calculations with LMDz-INCA-ORCHIDEE modelling system. Overall modelled  $O_3$  concentrations increased to 2050, but not as much when the  $CO_2$  effect is included.

At the European scale, studies with four CTMs found significant reductions in oxidized N concentrations and deposition over Europe between 2000-2050, reflecting anticipated future decreases in NO<sub>x</sub> emissions (Simpson et al., 2014). Much smaller changes (both increases and decreases) were found for reduced N deposition. These reflect the minor anticipated reduction in future European NH<sub>3</sub> emissions and the fact that the new climate effect on NH<sub>3</sub> emissions was not so far included in CTMs. The responses of the CTMs to the input emissions differed in some details, but overall the performance was similar across the different models.

Figure 7 shows the effects of the standard 2050 emissions and climate change effects on exceedance of critical-levels (CL) from the EMEP model. In this case, the figure also illustrates the estimated impact of the climate-induced increase in  $NH_3$  emissions discussed above. Although even a 30% increase in  $NH_3$  will not bring exceedances back to 2000s levels, such climate-induced increases in  $NH_3$  emissions cause CL exceedances that are substantially larger than those of the standard 2050 emission scenario.



**Fig. 7:** Frequency distribution of exceedances of the Critical Loads for eutrophying Nitrogen in Europe (EU28+). The red line (E05-M00-BC1) represents a year 2000 base-case and the E50-M50-BC3 scenario represents year 2050 with current emission estimates. The E50X20 and E50X30 scenarios illustrate calculations with 20% and 30% extra NH<sub>3</sub> emission due to climate-induced evaporation. See Simpson et al. (2014b) for more details.



**Fig. 8:** Calculated deposition of oxidised nitrogen (NO<sub>y</sub>) and reduced nitrogen (NH<sub>x</sub>), 1900-2050, with MATCH and EMEP MSC-W models. The climate effect on NH<sub>3</sub> emission is not included in these runs, which would further increase total NH<sub>x</sub> deposition by 2050 (cf. Fig. 7). (Engardt et al., 2015).

An important output from WP7 has been long-term simulations with the MATCH and EMEP models for the period 1900-2050 (**Figure 8**). The resulting fields of ozone and N-deposition from these models were also delivered to C2 and C4 partners as inputs to ecosystem models. These results, which show dramatic changes over the period, have been shown to compare rather well with historical observations of N-deposition. Deposition of NO<sub>y</sub> is predicted to fall significantly over the next 50 years, but NH<sub>x</sub> deposition remains high throughout the next 50 years. If the warming effect on NH<sub>3</sub> emission is included, NH<sub>x</sub> deposition will be even larger.

The EMEP MSC-W CTM was modified in order to take account of physical/chemical changes expected in the future, so that it is better able to predict air pollution metrics. The main modifications included the  $CO_2$  inhibition of isoprene emissions,  $CO_2$  inhibition of stomatal conductance, inclusion of ammonium-nitrate evaporation effect, addition of stress-induced BVOC to the model (Bergström et al., 2014, 2015), improved growing season estimates (Sakalli & Simpson, 2012), sensitive to temperature change, and various technical improvements to allow different types of climate model input. Calculations of  $O_3$  and N-deposition for 2050 have been carried out with the EMEP model using a range of climate effects, as have source-receptor calculations under different assumed climate impacts.

The new calculations with the 'climate-enhanced' EMEP model reinforce the message of the ECLAIRE ensemble and related studies, that emission changes are in general the key driver of future air pollution metrics. Although the use of, for example, a new photosynthesis module, or of  $CO_2$ -inhibition of isoprene emissions, modifies most air pollution

metrics to a certain extent, the changes are small compared with the effects mediated through emission changes. The most significant exception concerns metrics which are very sensitive to particular thresholds (e.g. POD<sub>3</sub>, a metric of phytotoxic ozone dose for crops).

A warmer climate would also have a range of other effects, such as changes in meteorological variables (water vapour, precipitation, drought; Simpson et al., 2014a), and likely increases in soil NO emissions. A warmer climate may also increase the evaporation of ammonium aerosol, leading to an increase in NH<sub>3</sub> concentrations and may also affect the atmospheric lifetime of ammonia due to changes in compensation points. Thus, even if emissions are the main driver of future air pollution metrics, climate will also have significant influence on the spatial pattern of O<sub>3</sub> levels and N deposition.

### Importance of local and regional variation

The aim of WP8 was to develop a better scientific understanding of the air pollution and climate change relationships at regional/local/landscape-scale and to develop sub-grid approaches for inclusion in large-scale models that enable a good representation of the multitude of processes that play a role on smaller scales.

Local scale interactions at spatial resolutions of  $1 \times 1, 5 \times 5$  and  $50 \times 50$  km<sup>2</sup> were investigated by means of the EMEP4UK model over Scotland and the Netherlands (**Figure 9**). The spatial distribution of the dry deposition of reduced nitrogen is highly dependent on the spatial distribution of ammonia emissions and, therefore, the model resolution. Although different, the total NL budget does not show a large difference between scales. The ammonia deposition velocity is relatively high and eventually most of the available ammonia (i.e. that not used to neutralise SO<sub>4</sub> or NO<sub>3</sub>) is dry-deposited within the NL domain. The differences in the NL budgets may be the result of resolving the national borders at the different resolutions.



**Fig. 9:** EMEP4UK modelled NH<sub>x</sub> dry deposition for the Dutch domain (mg N m<sup>-2</sup> for 2008). The black box shows an arbitrary 50 km × 50 km where the deposition is ~700 mg N m<sup>-2</sup>. The ranges of deposition in the same 50 km × 50 km grid square are: ~250 – 750 mg N m<sup>-2</sup> for the 5 km × 5 km resolution and ~250 - >850 mg N m<sup>-2</sup> for the 1 km × 1km resolution.



**Fig. 10:** Schematic showing the process of producing the sub-grid concentration predictions from short-range dispersion model simulations and high spatial resolution emission data.

WP8 also developed a parameterisation that can simulate the sub-grid spatial distributions of mean annual concentrations and deposition rates of air pollutants (specifically NH<sub>3</sub>, NO<sub>2</sub> and N deposition) within the grid cell of a large scale CTM (e.g. the EMEP MSC-W model) – see **Figure 10**. The resulting 'sub-grid distributions' provide an estimate of the spatial variability of the concentrations at 1 km resolution. A comparison of the modelled (sub-grid) concentrations with measured values shows that the modelled values compare reasonably with the measurements. Indeed, the sub-grid model for atmospheric concentrations seems to represent a substantial improvement on the predictions of the EMEP 50 km results for concentrations of NH<sub>3</sub> and NO<sub>2</sub>. The performance of the sub-grid model for wet deposition, however, is similar to that of the EMEP model. In all, the methodology shows promise and will be explored for routine application in future studies.

### 3.3. Component 3: Ecological Response Processes and Thresholds

Component 3 has improved our understanding of the effects of air pollutants, alone and in combination on terrestrial ecosystem functioning and services, and how these effects will be modified in a changing climate. The component comprised five inter-connected work packages (**Figure 1**) covering data mining and data re-use (WP9), experimental studies of effects and novel pollutant interactions (WPs10, 11) and modelling to determine novel thresholds (WP12) and ecosystem-scale impacts (WP13). All data mining, experiments and modelling focussed on realistic N deposition rates and O<sub>3</sub> or aerosol concentrations, reflecting current or predicted concentrations/deposition in the coming decades.

### Ozone pollution reduces carbon assimilation in many species

A data mining exercise was undertaken using a common methodology and template to extract data from the scientific literature on the effects of  $O_3$  on photosynthesis parameters in crops, grassland, heathland and wetland species (WP9). Using a combination of meta-analysis and mixed-effects modelling in R, responses were calculated as the percentage decrease in each parameter per ppb of  $O_3$  (**Table 1**). All effects of  $O_3$  were negative, with photosynthesis and stomatal conductance reduced by 0.33 to 0.40 % per ppb of  $O_3$ . An example of effects of

daylight  $O_3$  mean concentration on the net photosynthetic rate of cereals and non-cereal crops is given in **Figure 11**. These and other relationships were used in leaf- and plant-scale modelling in WP12 and ecosystem-scale modelling in WP13.

**Table 1:** Meta-analysis of effects of  $O_3$  on photosynthetic parameters of crops, heath, grassland and wetland species. The effect size is expressed as % change per ppb of  $O_3$ . 9999 iterations were run to calculate the 95% bootstrap confidence interval. The "\*" next to the effect size indicates at least a 95% confidence that it is not 0.

Process	Effect size	Bootstrap Cl
Biomass	-0.48*	-0.69 to -0.35
A <sub>sat</sub>	-0.33*	-0.47 to -0.18
J <sub>max</sub>	-0.15	-0.36 to 0.19
Net photosynthetic rate	-0.36*	-0.51 to -0.22
V <sub>cmax</sub>	-0.24*	-0.38 to -0.08
Stomatal conductance	-0.40*	-0.53 to -0.28

Note:  $A_{sat}$  is the photosynthetic rate at saturating light levels;  $J_{max}$  is the maximum rate of electron transport (an indication of the efficiency of the light capturing efficiency of photosynthesis);  $V_{cmax}$  is the maximum carboxylation velocity (an indication of the efficiency of the C fixation of photosynthesis).

Several experiments in WP10 included measurements of effects of  $O_3$  (with or without added N) on photosynthesis and biomass accumulation. In general, effects of  $O_3$  became more pronounced as the growing season progressed, in part reflecting the earlier senescence or die-back of leaves in elevated  $O_3$ . For example, elevated  $O_3$  (seasonal mean of 68 ppb) progressively reduced components of photosynthesis such as the maximum carboxylation velocity (V<sub>c max</sub>) in the tree species silver birch by 6% in June, 25% in July and 39% in September relative to the control treatment (**Figure 12**). Negative  $O_3$  effects on photosynthesis were mirrored in negative effects on biomass accumulation as indicated by data mining (**Figure 11b**) and ECLAIRE experiments on species such as silver birch, hornbeam, annual pasture species (**Figure 13**), leafy vegetable crops, and barley.

Under WP12, a novel method for modelling effects of  $O_3$  on net annual increment of trees was developed to estimate effects on trees at any stage in their life time, based on response functions from experiments with young trees. Previous response functions were for effects on relative biomass of young trees (Büker et al., 2015). This new method allows effects on carbon sequestration to be estimated spatially for several tree species making use of national reporting of tree net annual increment to the UN Framework Convention on Climate Change, and includes response functions for minimum, average and maximum effects (**Table 2**). The response functions are based on the stomatal uptake of  $O_3$  taking into account the varying effects of climate, soil moisture, seasonal changes and  $O_3$  on the opening and closing of the leaf stomatal pores.



**Fig. 11:** Results of data mining for effects of  $O_3$  on (a) the photosynthetic rate of crops and (b) trees, grassland and wetland vegetation.



**Fig. 12:** Relative variation in the photosynthesis parameter,  $V_{cmax}$ , by N and O<sub>3</sub> treatment over the course of the summer for a deciduous tree species, silver Birch. Low and high N treatments were 10 and 70 kg/ha/yr, respectively, whilst low and high O<sub>3</sub> treatments were 35 and 70 ppb (24 hour mean). Data are means +/- standard error.





(b) hornbeam root biomass (Charcoal-filtered air (CF), Non-filtered (NF), elevated O<sub>3</sub> (OZ+ & OZ++); (c) above ground biomass of Mediterranean annual pastures (CF, NF, NF+20 ppb O<sub>3</sub> and NF + 40 ppb of O<sub>3</sub>), with no added N (N0), 20kg ha<sup>-1</sup> y<sup>-1</sup> added N (N20) and 40kg ha<sup>-1</sup> y<sup>-1</sup> added N (N40).

Receptor	PODy	Default	R2	Min	R2	Max	R2
Norway spruce and Scots pine	1	y = -0.0057x + 1.0015	0.72	y = -0.0053x + 1.0014	0.71	y = -0.0065x + 1.0019	0.72
Norway spruce	1	y = -0.0054x + 1.0002	0.56	y = -0.0051x + 1.0003	0.55	y = -0.0062x + 1.0000	0.56
Birch & Beech	1	y = -0.0093x + 0.9461	0.59	y = -0.0090x + 0.9464	0.58	y = -0.0101x + 0.9449	0.59
Oak	1	y = -0.0057x + 1.0167	0.75	y = -0.0052x + 1.0142	0.75	y = -0.0066x + 1.0212	0.75
Aleppo pine	1	y = -0.005x +0.9998	0.64	y = -0.0046x + 0.9989	0.64	y = -0.0058x + 1.0013	0.65

**Table 2:** Statistical details (regression equation and R<sup>2</sup>) of the Net Annual Increment (NAI) doseresponse relationships for a variety of European forest tree species. Response functions for the 'standard', 'minimum' and 'maximum' growth curves are shown.

### Ozone alters nutrient absorption, utilisation and efficiency

New analysis of published data from exposure experiments conducted on wheat has revealed that  $O_3$  reduces the efficiency of fertilizer inputs (WP9). The fraction of N, P and K added to wheat as a fertilizer that ends up in the grains is negatively affected by  $O_3$  (Figure 14a).

This means that plants are less able to utilize added nutrients under  $O_3$  exposure which is expected to mean more nutrient losses to water supplies and also has important implications for the combined effects of  $O_3$  and N pollutants at the ecosystem scale (see below). Other experiments showed that  $O_3$  also reduces the ability of legumes such as clover to fix nitrogen (Hewitt et al., 2014, 2015).

As well as impacting on the nutrient efficiency and N fixation,  $O_3$  also reduces the reabsorption of nutrients from the leaves into the over-wintering parts of plants at the end of the growing season. This effect was detected in data mining and confirmed by the ECLAIRE experiments. When all data were combined, there was a clear negative effect of  $O_3$  on nitrogen resorption (**Figure 14b**). As a consequence, more N is available in leaf litter for microbial decomposition in the soil, triggering changes in biogeochemical cycling (see later section).



**Fig. 14:** Effects of  $O_3$  on (a) relative N fertilizer efficiency in wheat and (b) the resorption of leaf N in trees before litterfall, where ECLAIRE results are compared with previous studies. Species are silver birch (solardomes; Uddling et al., 2006) and paper birch (Lindroth et al., 2001) (WP9 activity).

# Ozone reduces the growth enhancing effects of nitrogen, and this is driven by effects on photosynthesis

New interaction experiments conducted in WP10 provided insight into the combined effects of  $O_3$  and N on leaf processes and biomass production. Factorial experiments were conducted with two Mediterranean tree species, annual Mediterranean grassland species and silver birch involving 2 to 4 N treatments and 4 to 7  $O_3$  treatments. Since  $O_3$  reduces growth and nitrogen increases growth, many have suggested that their effects could cancel each other out. The ECLAIRE analysis indicates that the actual picture is more complex, with significant interactive effects. When results from experiments in WP10 were combined with mined data from WP9, there was a clear indication that at higher  $O_3$  concentrations in the range 60 - 95 ppb, the stimulating effect of increasing N on root biomass was completely lost (Figure 13a). This effect was evident in individual ECLAIRE experiments, for example in hornbeam (Figure 13b), and was also seen in the above ground biomass of annual Mediterranean pastures (Figure 13c; Calvete-Sogo et al., 2014) and total biomass of silver birch (Figure 15a).

At the leaf level, added N increased photosynthesis, but this effect was less pronounced at higher  $O_3$  (e.g. **Figure 12**). This interaction can be viewed in different ways. On the one hand, this can be seen as N partially alleviating the negative effects of  $O_3$  on photosynthesis. Alternatively, it can also be viewed as  $O_3$  decreasing the plants ability to utilize N inputs. **The** 

dynamics of combined effects of  $O_3$  and N on photosynthesis during the growing season were included in the further development of the photosynthesis-based DO<sub>3</sub>SE model of  $O_3$  uptake and C assimilation (WP12). The new model, DO<sub>3</sub>SE-C, was able to reproduce the response of biomass to the combined effects of  $O_3$  and N deposition in ECLAIRE experiments (**Figure 15a,b**). In both experiments and modelling, the largest tree biomass occurred under situations with low  $O_3$  and high N whilst the lowest biomass occurred under high  $O_3$  and low N. In relative terms, the effect on biomass of increasing  $O_3$  under high N is greater than the effect under low N. In practice this means that the anticipated growth response to fertilizer inputs characteristic of clean conditions is not achieved at higher ambient  $O_3$  concentrations, with major implications for agricultural productivity and losses of other forms of nitrogen pollution.





### Combined effects of ozone and N at the ecosystem scale are not additive

The dynamics of the impacts of  $O_3$  and N on ecosystems were studied in several ways in the ECLAIRE project, including experimental investigations on grassland ecosystem processes, multi-factorial analysis of long-term forest ecosystems and modelling of temporal changes in greenhouse gas emissions and soil chemistry.

ECLAIRE provided funding in WP11 for additional measurements and data analysis for the final phase of a seven-year free air exposure of *Geo-Montani-Nardetum* subalpine grassland (2000 m.a.s.l.) in Switzerland to three O<sub>3</sub> concentrations and five N deposition rates in a cross-factorial design (Bassin et al., 2013). This high altitude site has a low background N deposition of ca. 4 kg N ha<sup>-1</sup> y<sup>-1</sup> and relatively high growing season mean O<sub>3</sub> concentration of 45 - 47 ppb. Under these high O<sub>3</sub> /low N and climatically challenging montane conditions, added N caused large changes in the community composition, with sedges becoming particularly dominant, whilst added O<sub>3</sub> at 1.2 and 1.6 x ambient had no effect on functional group composition and few effects on productivity; there were no significant O<sub>3</sub> x N interactions (Bassin et al., 2013). The lack of sensitivity to O<sub>3</sub> was attributed to enhanced levels of anti-oxidants for tolerance of UV radiation at high altitude, continual exposure to high background rather than peak O<sub>3</sub> and enhanced natural resilience in this long-lived, slow-growing community.

A separate field-scale exposure experiment was conducted in the UK in WP11 in which coastal grassland swards from 7 sites with similar precipitation, soil pH and vegetation type but varying

in their historical N deposition from 5.4 kg ha<sup>-1</sup> yr<sup>-1</sup> to 26 kg ha<sup>-1</sup> yr<sup>-1</sup> were exposed to ambient (mean 28 ppb), medium (mean 36 ppb) and high (mean 48 ppb)  $O_3$  in the Free Air  $O_3$  Exposure facility at NERC-Bangor. Long-term N deposition decreased total species richness, whilst many species showed increased leaf injury and/or accelerated leaf senescence with increasing  $O_3$  treatment (see ECLAIRE Third Periodic Report). In addition, the dissolved organic carbon (DOC) content of the water samples increased with increasing N and decreased with increasing  $O_3$ , probably corresponding to mesocosm productivity.

Fifteen years (1995 to 2010) of growth and deposition data from the LRTAP Convention's ICP Forests Europe-wide tree monitoring programme was analysed in WP9. It was found that relative forest increment increased up to a threshold of ca. 30 kg ha<sup>-1</sup> y<sup>-1</sup> of N, and levelled off at higher N levels. In general, deciduous forests were growing in areas with higher POD<sub>1</sub> and N deposition values (17-21 kg N ha<sup>-1</sup> yr<sup>-1</sup>, 34-36 mmol POD<sub>1</sub> m<sup>-2</sup>), where POD<sub>1</sub> is the accumulated phytotoxic O<sub>3</sub> dose over a threshold flux of 1 nmol m<sup>-2</sup>, than coniferous forests (11-15 kg N ha<sup>-1</sup> yr<sup>-1</sup>, 23 mmol POD<sub>1</sub> m<sup>-2</sup>). For coniferous forests, POD<sub>1</sub> and N deposition were strongly positively non-linearly related, making it difficult to disentangle the direct impact of N deposition and POD on growth. Thus, **the negative impact of POD<sub>1</sub> on forest growth was masked by the positive effect of N deposition and temperature on forest growth. However, at N saturated plots POD<sub>1</sub> showed a clear negative correlation to forest increment** with a 2% decrease of forest increment per 1 mmol m<sup>-2</sup> POD<sub>1</sub>.

To gain further understanding of the combined effects of O<sub>3</sub> and N on ecosystems and predict long-term changes over the coming decades, the MADOC model of N and acidity effects on vegetation growth, soil organic matter turnover, acid-base dynamics, and organic matter mobility (Rowe et al., 2014) was extended in WP13 to include  $O_3$  and  $O_3$  - N interactions. Following an extensive review of the literature on  $O_3$  effects and  $O_3$  - N interactions (WP9), strong and consistent evidence was found for two key ecosystem-scale effects: a reduction in Net Primary Productivity (NPP) and early leaf senescence, resulting in reduced resorption of N and a greater concentration of N in leaf litter. These effects were incorporated into the MADOC model using NPP reduction functions derived from the scientific literature in WP9 for wet grassland and bog, other grassland and woodland (Figure 12b). Site-specific date were collected from long-term measurement and/or manipulation sites as part of WP9 and used within MADOC for explorations of ecosystem sensitivity to combined air pollution and climate change drivers. Scenarios applied (from 2020) were increases in mean annual temperature of 2 and 4 °C, and increases and/or decreases in N and O<sub>3</sub> pollution by +/- 20%. Simulations with MADOC showed broadly opposing responses to O<sub>3</sub> and N pollution at the ecosystem scale. In general, additional N deposition increases the amount of available N within the ecosystem, which in turn stimulates productivity in N-limited semi-natural ecosystems, as shown in Figure 16. These modelled increases in NPP have a cascading effect on other ecosystem properties and functions, for example leading to an increase in soil C (implying an increase in CO<sub>2</sub> sequestration) and increasing leaching of nitrate and DOC to surface waters. Although the magnitude of these effects are predicted to vary between ecosystems (compare left and right panels of Figure 16) the general direction of change is predicted to be consistent.

### Aerosols damage stomatal functioning and reduce plant resistance to drought

Experiments were conducted in WP11 to determine the effects of hygroscopic particles on leaves from aerosol and trace gas deposition on stomatal control of water loss from leaves. It

was found that ambient concentrations of aerosols depositing on leaves can reduce water use efficiency of plants (Figure 17). The particles provide a wick mechanism that increases stomatal conductance of water under conditions of low soil moisture availability. This effect is particularly deleterious for those species that are less able to respond to drying soils by closing stomata when soil moisture is limited. Plant species that have a morphology that is most efficient at capturing particles and/or long-lived leaves/needles are at greatest risk from this effect. The experimental measurements in ECLAIRE have allowed a first dose-response parametrization of this effect to be established, which will be exploited after the project in testing the implications in DGVMs.



**Fig. 16:** MADOC scenario assessment for the effects of changing future N and  $O_3$  on simulations of soil available N, net primary production, soil carbon and leachate dissolved organic carbon for the Gårdsjön forest experimental site (left) and Whim Bog experiment (right). Red, black and blue lines represent high, business as usual and low N deposition scenarios; short-dashed, solid and long-dashed lines represent high, business as usual and low  $O_3$  scenarios.



Fig. 17: Effects of aerosols on the minimum stomatal conductance of leaves

# Climate change will modify responses of vegetation and ecosystems to pollutants

A key objective of ECLAIRE has been to gain further insight into the combined effects of pollutants under a changing climate. In C3, this was addressed via data mining, experiments and modelling, including through the examples provided above. Additional climate change – interaction experiments were conducted for the crop barley under controlled climatic conditions and for dry heathland under field conditions (WP10). Experiments examined the response of grain weight in over 100 genotypes of barley. Grain weight decreased with elevated temperature and  $O_3$ , and increased with elevated CO<sub>2</sub> (Ingvordsen et al., 2015).

Long-term ecosystem experiments with dry heathland have demonstrated that **drought**, **progressive N dilution and non-linear effects between climate change factors can reduce the effect of CO**<sub>2</sub> **in stimulating photosynthesis.** It was found that **adding O**<sub>3</sub> **leads to even more negative effects on photosynthesis than when plants were acclimated to long term elevated CO**<sub>2</sub> **and drought**. In a different study the effects of climate on inter-annual variability of annual Mediterranean pastures was analysed in WP10 in relation to O<sub>3</sub> sensitivity. In dry years, there was a very high proportion of less O<sub>3</sub>-sensitive grass **species whilst in moist years**, **O**<sub>3</sub>-sensitive herbs, particularly legumes dominated in **Mediterranean pastures**. Ozone fluxes were also lower in the dry than wetter years; if drier years become more prevalent as predicted under climate change, reduced O<sub>3</sub> effects will be offset by reductions in biomass and nutritive quality.

These results can be seen in combination with the finding that  $O_3$  effects are largest at high N availability (previous sections). They paint a picture where  $O_3$  can have its largest relative effects under otherwise optimal conditions of good nutrient and water availability.

A combination of meta-analysis of published data and measurements of biogenic volatile organic compounds (BVOCs) within and above forest canopies and under experimental O<sub>3</sub> and N combinations was undertaken in order to gain further insight into the many factors that influence their emissions from plants. These are important in a changing climate because, depending on their chemical composition and presence/absence of other O<sub>3</sub> precursors they can lead to either the removal or enhanced formation of O<sub>3</sub>. It was found, for example, that **isoprenoid emissions increase with increasing temperature, and decrease with increasing CO<sub>2</sub> and soil water stress (Figure 18). Effects of O<sub>3</sub> and N, single and in combination, were inconsistent in the scientific literature and new ECLAIRE measurements** 

were made to provide further insight. In silver birch exposed to  $O_3$  (Figure 15),  $O_3$  exposure increased BVOC emissions, whilst N treatment decreased emissions of some BVOCs such as  $\alpha$ -pinene and  $\beta$ -pinene but increased emissions of others (data not presented). It was concluded that  $O_3$  and N pollution have the potential to affect global BVOC via direct effects on plant emission rates and changes in leaf area.

Field measurements in a Mediterranean Holm oak forest showed that  $O_3$  fluxes are highest during the central hours of warm days. This is due to both stomatal uptake of  $O_3$  into the leaves and non-stomatal deposition of  $O_3$  to leaf surfaces and via chemical reactions with monoterpenes and isoprenes released from the leaves of Holm oak during these climatic conditions. Low temperatures lead to almost negligible BVOC fluxes during the winter reducing non-stomatal sinks for  $O_3$ . In addition, during the day  $NO_2$  formed and was deposited to the Holm oak canopy whilst at night  $O_3$  was completely scavenged below the canopy by NO.



**Fig. 18:** Percent change in isoprene and total monoterpenes emission under the effect of different climate change drivers. Symbols are bracketed by 95% bootstrapped confidence intervals. Mean level of stress and number of observations (in parenthesis) are also given.

### 3.4. Component 4: Ecological responses at regional & EU scales

Component 4 has enhanced our insight into how the changes in air pollutants in interaction with climate change will affect terrestrial ecosystem functioning and services. The component comprised four inter-connected work packages with the objective to assess for terrestrial ecosystems the: (i) effects of combined air pollution and climate change scenarios on terrestrial productivity and ecosystem carbon exchange (WP14), (ii) soil quality and plant species diversity under different air pollution and climate scenarios for forests and semi-natural systems (WP15) with related critical thresholds for nitrogen deposition and ozone uptake and their exceedances (WP16). Furthermore, the uncertainty in critical N thresholds and their exceedances has been evaluated based on model simulations at several grid resolutions at national scale and landscape scale (WP17).

### Joint effect of N and O<sub>3</sub> varies for net primary production & evapotranspiration

An ensemble application and inter-comparison of the results of four models (CLM, O-CN, Jules and LPJ-Guess) on the long-term impacts of various scenarios of climate change, air quality change (exposure to  $O_3$  and  $CO_2$  and deposition of nitrogen) on net primary production

evapotranspiration and water use efficiency of forests and semi-natural systems, has been carried out (Cescatti et al., in prep.). All models show the positive effect of N deposition and the negative effect of  $O_3$  on NPP but the joint effect of N and  $O_3$  together differs for the various models. Model results indicate that N does not increase NPP if there is  $O_3$ , while  $O_3$  reduces NPP whether there is N or not, indicating that effects of N and  $O_3$  are not additive. The effects of N and  $O_3$  on evapotranspiration are largely the same as for NPP. N increases and  $O_3$  decreases evapotranspiration. N and  $O_3$  together reduce evapotranspiration, implying that the effect of  $O_3$  seems stronger than the effect of N on this indicator.

### Tree carbon sequestration is much larger than soil carbon sequestration

An empirical soil-based model called EUgrow-VSD+ was extended and applied to assess interactions and long-term impacts of climate change and air quality on forest carbon sequestration (De Vries et al., in prep.). The model includes empirical response functions between net primary production and temperature, water availability,  $CO_2$  concentration, N deposition and  $O_3$  exposure, in terms of phytotoxic ozone dose (POD). Results show that the ongoing tree C sequestration is much larger (above 1000 kg C ha<sup>-1</sup> yr<sup>-1</sup>) than soil C sequestration (between -20-20 kg C ha<sup>-1</sup> yr<sup>-1</sup>). The soil C pool changes reflect on average the changes in tree C pools as this affects the C input by litter-fall. However, unlike tree C sequestration, the changes can be negative since soil respiration can be higher than litter C input. The decrease in C sequestration in the period after 2005 to negative values in 2050 for all scenarios is most likely due to climate change, on average increasing soil respiration by an increased temperature. Results show a rather strong impact of the growth responses to N deposition (linear or non-linear) and an even larger impact of the two empirical  $O_3$  exposure functions, considering either total biomass or net annual increment (Figure 19).



**Fig. 19**: Temporal development of European average carbon sequestration in trees (left) and in soil (B) in response to changes in climate,  $CO_2$  concentration, N deposition and  $O_3$  exposure for two growth responses to N deposition (linear and non-linear) and  $O_3$  exposure (total biomass and net annual increment, NAI). (Based on de Vries et al., in prep.)

# Impacts of drivers of forest production and tree carbon sequestration are dependent on the status of other drivers

Impacts of individual drivers on forest production and tree carbon sequestration are dependent on the status of other drivers, as illustrated in **Figure 20**. This shows the results of the empirical EUgrow-VSD+ model. **The model predicts that the fertilizing CO<sub>2</sub> effect is higher at elevated N, than at low N.** Similarly, the model predicts that the fertilizing effect of elevated N availability is higher at elevated CO<sub>2</sub> than at low CO<sub>2</sub>. In general, O<sub>3</sub> impacts appear to be independent of the other drivers. Climate impacts in relation to other driver are highly site specific and results are not trivial to explain at a European wide scale. **In the past the most**  important driver for growth change has been N deposition and in the future it is expected to be the combination of climate and CO<sub>2</sub>, in combination with the reduction in ozone impacts. The latter is due to a predicted decrease in phytotoxic ozone dose (POD), which largely compensates for the reduced fertilizing effect of N due to reduced N deposition.

### Tree and soil carbon sequestration is expected to increase in central Europe, but not in Northern and southern Europe

Spatial patterns for the time-averaged tree and soil carbon sequestration for the period 1900-1950, 1950- 2000 and 2000- 2050 based on the empirical EUgrow-VSD+ model are given in **Figure 21**, for the reference model, including interactions between drivers.



**Fig. 20**: Influence of the single drivers computed, using the linear N model and the total biomass  $O_3$  response model (Based on de Vries et al., in prep.).



**Fig. 21:** Spatial variation in calculated tree C sequestration over Europe in the period 1900-1950, 1950-2000 and 2000-2050, using the reference model with a linear N response model and a total biomass response to POD (Based on de Vries et al., in prep.).



**Fig. 22:** The spatial distribution of the habitat suitability index of EUNIS classes in EMEP grid cells in 2005 (Left) and right the calculated increase in HSI between 1990 and 2050; median per grid cell in response to the ECLAIRE scenario (right). (Based on Reinds et al., in prep.)

Results show that the 50-year average C sequestration increases 1900-1950 < 1950-2000 < 2000-2050 in C. Europe, but not in N. and S. Europe (**Figure 21**). In these regions, the growth rate stays rather constant. Apparently, water availability limitations mainly offset the effects of CO<sub>2</sub> and temperature increase in Southern Europe, whereas limitations due to N availability and O<sub>3</sub> exposure seem to offset those effects in Northern Europe.

## Future climate change affects plant species diversity more than planned future reductions in N deposition

Both N deposition and climate change affects plant species diversity (De Vries et al., 2015; Hettelingh et al., 2015; Rowe et al., 2015; Van Dobben et al., 2015). An empirical model, called PROPS, has been developed that describes occurrence probability functions for about 4000 European plant species as a function of abiotic conditions (pH, soil C/N ratio, N deposition, yearly precipitation and yearly average temperature) based on an existing database of about 800000 relevés (Wamelink et al., in prep.). Application of the PROPS model combined with the dynamic soil model VSD+ to Europe in **ECLAIRE showed a stronger influence of climate and a lower impact of N deposition change on the computed change in a habitat suitability index, HSI (Figure 22). This suggests that climate change results in a stronger loss of diversity than expected future reductions in N deposition.** 



**Fig. 23:** Exceedance of CLs for biodiversity under 2010 (left) and 2050 (right) N and S deposition (Based on Posch et al., in prep.).

# A new way of looking at N and S critical loads indicates different spatial patterns of adverse effects on species diversity.

In an exploratory approach developed within ECLAIRE, the above mentioned plant species response curves and the characterisation of every (European) habitat by a number of 'typical' species, allowed new values of critical (or rather 'optimal') loads of N- and S-deposition to be derived. These are based on an agreed threshold value of the habitat suitability index (HSI). The 'average accumulated exceedance' (AAE) of thus derived biodiversity based critical loads for N and S show relatively limited exceedances for the years 2010 and even lower exceedances are predicted for the year 2050 (Posch et al., in prep.). Results show that exceedances in the Netherlands and the Po area still stay in 2050, even after the predicted N deposition reduction (**Figure 23**). These findings provide a new way of assessing the effects of N and S deposition on ecosystems that can stimulate future model discussion and refinement for cost-benefit analyses. The actual extent of exceedance in this new approach is dependent on parameter setting of the HSI that should be further evaluated in future.

# Exceedances of critical thresholds for ozone uptake have a significant impact on forest growth

The impact of current and expected phytotoxic ozone dose (POD) on reduction in the net annual increment (NAI) of forests has been assessed based on linear relationship between NAI and POD<sub>1</sub>, distinguishing Norway spruce, Scots pine, Other conifers, Beech/Birch, Oak and Other broadleaves, as derived under C3. Results indicate that current reductions in Net Annual Increment of the most sensitive species, i.e. birch, due to  $O_3$  vary from about 10-15% in Northern Europe to more than 30% in Central Europe, while estimated future reductions in 2050 are on average about 5% less (Figure 24). Note, however, that this is an estimate for birch which is the most sensitive species, while impacts on other tree species are substantially lower.



**Fig. 24:** Reductions in Net Annual Increment (NAI) for birch (*Betula* spp.) due to ambient O<sub>3</sub> exposure in the year 2000 (left) and 2050 (right).



**Fig. 25:** Average accumulated exceedances for  $50 \times 50 \text{ km}^2$  grid squares in the Netherlands (top) and Scotland (bottom) calculated using nitrogen deposition data at three model spatial resolutions (left:  $1 \times 1 \text{ km}^2$ ; centre:  $5 \times 5 \text{ km}^2$  and right:  $50 \times 50 \text{ km}^2$ ). Note: different exceedance range in Dutch and Scottish maps (Theobald et al., in prep.).

## There is a tendency to overestimate exceedance of nitrogen critical loads when using coarse resolution data

The impact of the used spatial resolution on critical N thresholds and their exceedances was assessed for the Netherlands and Scotland using the three different spatial resolutions of nitrogen deposition data, i.e.  $50 \times 50 \text{ km}^2$ , being the standard used at European scale and the much lower resolutions of  $5 \times 5 \text{ km}^2$  and  $1 \times 1 \text{ km}^2$ . **Results showed that using the coarse resolution data (50 x 50 km<sup>2</sup>) tend to overestimate average accumulated exceedance of critical nitrogen loads (Figure 25)**. Although there are small differences between the

individual AAE values for a particular 50  $\times$  50 km<sup>2</sup> grid square, the general pattern and range of exceedances slightly increases going from 1  $\times$  1 km<sup>2</sup> to 50  $\times$  50 km<sup>2</sup> in line with the calculated slightly increasing N deposition in this direction for both domains (on average from 21.8 - 22.4 kg N ha<sup>-1</sup> yr<sup>-1</sup> in the Netherlands and from 5.1 to 5.4 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Scotland) (Theobald et al., in prep.).

### 3.5. Component 5: Integrated Risk Assessment and Policy Tools

The ECLAIRE project has provide new information for policy assessments through targeted studies on the interaction between ozone damage, nitrogen impacts on biodiversity and on the potential alleviation of ozone damage by adding nitrogen. All of these activities focused on re-evaluating current policy responses and recommendations on abatement measures. As climate change may affect several of the relationships contributing to any of the impacts discussed, ECLAIRE also addressed the question if these policy-relevant recommendations might be considered robust and also valid under climate change conditions, or if there might be situations of measures turning to become disadvantageous in the future.

### Role for Nitrogen compounds in carbon sequestration

Additional nitrogen available boosts forest growth. Especially in nitrogen-limited boreal forests, which constitute a significant fraction of forested area in Europe and in the EU, increased anthropogenic air pollution will allow forests to grow more quickly, contributing to enhanced wood production. However, for the more densely populated parts of Europe, continuous long-term pollution deposition has led to demonstrated decreases of forest growth. Here also emissions and deposition of nitrogen compounds (oxidized and reduced nitrogen) occur at a larger rate. Studies in Switzerland and in Belgium have shown such negative impacts in biomass accumulation, which in at least the latter case have been attributed to the need of growing forest biomass to extract phosphorous from soil. In the long term, P deficiency occurs in soils impeding current growth. Also other causes for growth reductions have been discussed, such as ammonium accumulation having negative impacts on nutrient balances, mycorrhiza composition and ground vegetation (Bobbink and Hettelingh, 2011).

**Figure 26** displays the response of N retained and of biomass growth as a function of atmospheric deposition of nitrogen. At levels below  $N_1$  nitrogen is effectively absorbed by biomass, and not released to the environment, while N increases the rate of plant growth. Above that level, nitrogen starts to leak, affecting affects other environmental pools. Growth still increases but arrives at a maximum level  $N_2$ , beyond which addition of nitrogen leads to a decline of growth, which above  $N_3$  becomes even lower than an unaffected "natural" system.



**Fig. 26:** Schematized nonlinear responses of (a) nitrogen retention efficiency and (b) net primary production (NPP) to external of N inputs to forests (de Vries et al., 2014).

For forest ecosystems, the level of N deposition at which growth is impaired ( $N_2$ ) has been found experimentally to be as low as 15 kg N/ha/year (see Braun et al., 2010; Kint et al., 2012, and see ECLAIRE deliverable D20.7 for more details). This level must not be understood as conflicting with any lower thresholds implemented to protect other ecosystem-relevant parameters (e.g. biodiversity change). As indicated in the left side of **Figure 26**, the system leaks nitrogen even before arriving at point  $N_2$ . This release may give rise to soil acidification and/or to eutrophication, for which critical loads may be even smaller than those needed to protect forest growth.

### Ozone

Results of the GAINS model derived in the framework of the project demonstrate only limited potential of emission reductions to further reduce vegetation exposure to ozone. Significant improvements have been seen in the past, which were due to reductions of emissions of  $NO_{x_1}$ for example achieved by introducing the 3-way catalyst in gasoline-driven vehicles, or reducing emissions of Volatile Organic Compounds from the use of solvents in industry. However, the technical potential for further improvements beyond the current legislation already implemented is currently rather limited. This includes expected reductions in NO<sub>x</sub> emissions from diesel engines, which have not yet been fully delivered by car manufacturers. Although further reductions in NO<sub>x</sub> and VOCs under proposals for the NECD will give major benefits for human health by reducing O<sub>3</sub> concentrations, the ecosystem benefits of will be much be more limited. The main reasons for this are the high contribution of midrange ozone concentrations to exposure metrics (as ozone flux) resulting in rather small sensitivity to reductions in European emissions, and the large contribution of the hemispheric background to European mid-range O<sub>3</sub> levels. To make substantial progress in reducing the  $O_3$  threat to European ecosystems, further efforts will also be needed to reduce  $O_3$ precursors (especially methane) from non-European sources. New technological development may also help go further with  $NO_x$  in the future (**Box 1**).

Figure 27 presents these O<sub>3</sub> results in detail, with data deriving from a number of studies compiled for ECLAIRE. In addition to ozone fluxes (as POD values), the figure also includes the SOMO35 metric (Sum of Ozone Means Over 35 ppb), which is relevant for human health protection. All data are given relative to 2010 and in relation to the hemispheric background situation. It becomes evident that the O<sub>3</sub> flux endpoints (POD<sub>1</sub> or POD<sub>3</sub>) become virtually undistinguishable on a relative scale, and also that it is a less sensitive parameter, compared to e.g. SOMO35: strong changes in input (emissions) will result in only little effective variation. Nevertheless, results show a clear decrease between 1990 and 2010, despite of the increasing hemispheric background and (economic) activity which would have triggered further increase towards the "hypothetical 2010" markers. Further improvements may be expected under "current legislation" for the year 2030. Using the technological optimum of emission abatement measures implemented in the GAINS model, in 2030 the Maximum Technically Feasible Reductions (MTFR) may be achievable. This technological limit, which comes at considerable abatement costs, shows rather little difference from the expected 2030 situation - much less than what has been achieved between the "hypothetical" and the actual 2010 results. Note that the EU Commission's 2014 Clean Air proposal for human health protection (see Amann et al., 2014 for details) comes up with clearly lower ambitions, which would result only in a quarter of the MTFR achievements. Returning to the 1990's hemispheric ozone background would allow arriving at twice the positive effects on reducing ozone damage to vegetation compared to the Commission proposal.



**Fig. 27:** Developments of relative phytotoxic ozone dose (POD) and SOMO35 (Sum of Ozone Means Over 35 ppb, related to human health) changes in the past (starting from a lower hemispheric background) and expectations under current legislation as well as potential for the year 2030.

In consequence, further reductions of emissions from European sources are unlikely to be effective in reducing plant exposure to ozone by 2030. Instead, improvement of the hemispheric background situation (much of it via reducing  $CH_4$  emissions) seems to be able to further reduce  $O_3$  damage to plants in Europe, while simultaneously improving the air quality situation in India, China or in North America. Here the hemispheric interactions allow to establish positive results across the continents, assuming that air pollution measures are properly implemented.

#### BOX 1: New thinking for future NO<sub>x</sub> control technologies

In the UNEP Global Overview on Nutrient Management "Our Nutrient World" to which ECLAIRE contributed, it was identified that there are also opportunities for new technical approaches to reduce  $NO_x$  emissions (Sutton et al., 2013). Current technologies have so far focused on denitrification of  $NO_x$  to form di-nitrogen (N<sub>2</sub>). As much has been done already, going further becomes increasingly expensive.

Among 10 key actions for better nutrient management, "Our Nutrient World" identified that insufficient investment had so far been placed in new technologies for "NO<sub>x</sub> Capture and Utilization" (NCU), whereby NO<sub>x</sub> is converted to nitrate (NO<sub>3</sub><sup>-</sup>) allowing it to be used for fertilizer and other product manufacture.

Globally, 40 Gg of nitrogen is emitted to the atmosphere annually as  $NO_x$ , having a fertilizer value of around  $\Subset 38$  billion per year. This points to a major financial opportunity for such Circular Economy thinking to stimulate new technology development for  $NO_x$  reduction in the future. Until now, insignificant investment has been made in NCU technologies, where the starting point would be large combustion plants.

### Ozone and nitrogen interaction

Both ozone and deposited nitrogen are deeply entangled in the photosynthesis process with major effects on biomass formation. The mechanisms include stomatal entry and disruption of physiological processes for  $O_3$  and complex stimulation of nitrogen cycling processes in plants and soil. These mechanisms have been investigated in ECLAIRE and the current status is described in detail elsewhere (ECLAIRE deliverable D12.3) by considering effects on net assimilation (A<sub>n</sub>) and stomatal conductance (g<sub>sto</sub>) that have been incorporated into a new A<sub>n</sub>-g<sub>sto</sub> version of the DO3SE model. This has established new dose-response relationships between  $O_3$  and annual biomass increments in forests in the context of changing N impacts.

In essence, the interaction may be regarded in two contrasting ways. **Ozone can be seen to impede the fertilizer effect of N, but likewise the addition of N can be considered a way to partially alleviate ozone damage.** The dose-response relationships (cf. **Figure 15b**) suggest that, under any given level of available N, POD will have a consistent and rather constant negative effect on biomass increments. Stark differences are to be seen between different plants (deciduous vs. coniferous trees). Moreover, at higher levels of N deposition (again, depending on tree species: a generalized level would be around ~30 kg/ha N) leaf N concentrations (and hence effects on photosynthesis) will level off as a function of deposition. Hence the beneficial effects of adding N to compensate negative impacts of O<sub>3</sub> will be lost at such high deposition rates.

Further processing of these findings of ECLAIRE will continue after the formal end of the project to further challenge the relationships and deepen understanding of these insights. For example, some ECLAIRE experiments showed that  $O_3$  had a larger effect at high N deposition, implying that  $O_3$  reduced the ability of plants to utilize nitrogen, leading to reduced production and a cascade of other N effects (N leaching and N<sub>2</sub>O emissions).

### **Ecosystems services**

Work in ECLAIRE has demonstrated that, while effects of air pollution on ecosystems are evident, quantification in monetary terms has proved to be challenging. This information is nevertheless important as it provides an input to cost-benefit analysis. For some services provision of data for cost-benefit analysis is straightforward. Examples include the relationships of forest productivity as a function of temperature change, CO<sub>2</sub>- and N-fertilization. However, many other possible services can be identified only, but robust quantification of costs remains difficult. Examples could the cultural and amenity value of healthy ecosystems and protected habitats. Neglecting these other relationships would imply to set the effects to zero, which is clearly unreasonable. Therefore ECLAIRE has put particular effort to find alternatives approaches to overcome these issues.

The following approaches to valuing the ecosystem effects of air pollution have been investigated in ECLAIRE (e.g. Maas, 2014):

- Quantification of marketed ecosystem services
- Estimation of 'willingness-to-pay' for non-marketed services
- Estimation of ecosystem restoration costs
- Consideration of elimination costs (i.e., regulatory revealed preference)
- Estimating the cost implication of an existing legal requirement for conservation
- Consideration of a Nitrogen Use Efficiency approach

In the last case, valuation is based on the amount of fertilizer saved under improved N use efficiency. While only the first and the last approaches listed above give market based figures, a comparison of three independent approaches converged towards broadly similar values. It was found that approaches based on restoration costs, willingness-to-pay and elimination (regulatory revealed preference) costs all led to a benefit/cost ratio for air pollution mitigation much lower for biodiversity protection than for health protection.

The approach based on the existing legal requirement for habitat conservation also provided informative. In this case it can be considered that a decision has already been taken by society in protecting certain nature areas, such as the "Natura 2000" areas. Legal obligations exist demanding "no net loss of biodiversity" from these areas in the EU28. **Based on EU nature legislation, a firm and consistent guidance to air pollution impacts can be developed. This requires to establish "biodiversity" as an endpoint in the GAINS system,** using atmospheric emissions and abatement strategies as an input. The underlying assumptions and strategies are described in ECLAIRE deliverable D19.4.

# ECLAIRE optimization scenario and biodiversity benefits from health-related measures

The impact of S and N deposition on the habitat suitability of vegetation classes has been used as a new indicator for biodiversity impacts of air pollution in Natura 2000 areas. Using this indicator, an illustrative optimization scenario ("ECLAIRE scenario") has been developed. This approach allows the advantages from a proposed health-related air pollution abatement package to be investigated with respect to biodiversity. The "cost curve" presented in **Figure 28** describes the cost of abatement measures (above the costs of current legislation, CLE) needed to arrive at a given target. In this metric, the target is a certain percentage of the difference between the CLE at 0 and the "maximum feasible reduction" (MFR) scenario at 100, taken from the total cumulated threshold exceedance of all protected areas in the EU.

As shown in **Figure 28**, just implementing the health-related elements of the Air Quality Package proposed by the European Commission (COM) (see Amann et al., 2015) will take care of 71.2% of the maximum achievable by technical measures. According to this approach, no specific consideration on biodiversity needs to be taken as emission abatement measures are largely the same. Note that originally the COM proposal was defined to take care of 67% of the potential to mitigate "Years of life lost".

In order to demonstrate the effect of a marginal change, the ECLAIRE scenario optimizes abatement measures at a level that just slightly exceeds those of the commission proposal, while maintaining the health target of 67% it simultaneously increases the biodiversity gap closure to 75%. The average accumulated exceedance (per ha of protected area) decreases, from the CLE case roughly to one half of this value, while the area Natura 2000 sites that are exceeded decreases by a quarter. Compared with what had been achieved already in the European Commission proposal, the ECLAIRE scenario reduces exceedance by a further 4%, while reducing the area of Natura 2000 sites exceeded by 1%. In order to achieve these improvements, additional costs extend to just 23 M€ or only 1.1% of the costs assigned to the European Commission proposal (additional to those already spent for the CLE scenario). This demonstrates the potentials of combined treatment of health and biodiversity protection.



**Fig. 28:** GAINS cost curve for optimizing European air pollution abatement, starting from baseline (blue) vs. from the European Commission Air Quality Package proposal in relation to human health protection goals (green). The illustrative ECLAIRE scenario developed here is marked by the red asterisk.

### Impacts of a future climate

Several of the relationships described by the GAINS model are affected by climate, most prominently by ambient temperature and by humidity/precipitation. With an increasing understanding of the future climate impacts in 2050 and in a more distant future (here termed the "nominal 2100 scenario"), it becomes relevant to shed light also on the effects a changed climate may have on vegetation response to air pollution.

There are several parameters impacting on the emissions from ecosystems, with higher temperatures in general tending to increase them. But also the sensitivity of ecosystems to air pollutants may be affected. With regard to the biodiversity indicator developed in ECLAIRE, critical loads have been assessed for the conditions of a climate scenario representing 2050 as well as 2100. An overall increase in sensitivity can be demonstrated, i.e. the same level of negative effects already appears at lower levels of emissions. Additional efforts here need to be regarded as climate adaptation measures. The extent of such adaptation may be assessed from **Table 3**, where we investigate impacts of an increased temperature for 2050 on climate change due to altered  $NH_3$  emissions as well as due to changed vegetation impacts.

In the analysis shown in **Table 3**, the impact of revised sensitivity of biodiversity and of increased NH<sub>3</sub> emissions are calculated for 2050, but for the matter of just determining adaptation costs, anthropogenic activities and implemented technologies are kept at a 2030 CLE situation as the central case. Results indicate (i) that the effect of climate on the sensitivity of biodiversity is even larger than that of increased ammonia emissions (at least for the case investigated) and (ii) that adaptation is readily available at low costs in the CLE case, but may become quite costly once applied on existing abatement strategies. In those cases, the cheap options have been taken already which limits further

possibility of low-cost abatement. Only the additional costs under CLE are in the range of the additional costs created by the ECLAIRE scenario alone (23 M€/yr above the COM proposal.

	Central case, Current climate 2030		With biodiversity indicators under climate change**)			With higher NH₃ emissions due to climate change ***)		
	HS		HS indi∙	Additional	costs to	HS indi-	Additional	costs to
	indicator	Costs *)	cator	return to cer	tral case	cator	return to	central
	(eq/ha)		(eq/ha)	*)		(eq/ha)	case	e *)
CLE	26.0	0	30.9	+95	0.11%	26.7	+26	0.03%
COM proposal	14.6	2189	17.7	+889	1.03%	14.9	+236	0.27%
ECLAIRE scenario	14.0	2212	16.3	+1333	1.54%	14.4	+386	0.33%

**Table 3:** Costs to compensate increased biodiversity impacts caused by climate change comparing current legislation (CLE) the Air Quality Package proposal of the European Commission (COM) and the ECLAIRE scenario focused on 2050. HS refers to critical loads exceedance according to the Habitat Suitability Indicator developed in ECLAIRE (see Component 4).

\*) all costs in M∉yr for the EU-28 on top of current legislation (CLE), expressed as % of CLE costs.

\*\*) for 2050 climate scenario (~1° higher temperature).

\*\*\*) 4% increased total NH<sub>3</sub> emissions in EU-28. In this case a smaller estimated effect is applied than estimated elsewhere in this report to account for the potential of adaptive practices by farmers. Based on Sutton et al. (2013, Eq. S3) a 1° C increase would raise NH<sub>3</sub> emissions by 7% (uncertainty range: 4-11%), excluding the possible moderating effects of adaptive practices.

One of the uncertainties in this comparison is the extent to which NH<sub>3</sub> emissions will increase in a warmer climate. Based on chemical thermodynamics alone, the Q<sub>10</sub> would be 3-4, though trade-offs with other processes led Sutton et al. (2013) to adopt a smaller Q<sub>10</sub> of 2 (1.5-3). For a 1 °C warming (indicative for 2050) these imply NH<sub>3</sub> emission increases of 13% (thermodynamics) or 4 (7-11)% (Sutton et al.). In **Table 3**, a value equivalent to the bottom of this range was used to account for possible adaptive practices by farmers and the use of controlled environment animal housing. However, even if the mid-range dependence were used (7% increase per °C), so that the additional critical load exceedance and costs roughly doubled, the estimated effect of climate on the biodiversity indicators would still larger than the NH<sub>3</sub> effect. Only in the case of the upper temperature sensitivity to NH<sub>3</sub> (11% increase per °C) would the two effects be comparable in magnitude according to this assessment. It should be emphasized that these values are based on the 2050 scenario. **The changes for 2100 (c.** 4°C **increase compared with present) would give additional exceedances and costs of at least four times these values, due to the non-linearity of the relationships.** 

These results point to the continued need to assess and evaluate ecosystem and biodiversity damage due to air pollution. With adaptation costs increasing strikingly with ambition to maintain vegetation and its functions, an abatement regime will need to be pursued actively in the long-term. These outcomes provide an indication of the extra efforts that would be needed if further progress with the ecosystem goals of the revised National Emissions Ceilings Directive and its successors is to be achieved by 2050 and beyond.

The results show that climate change is increasing the costs of air pollution mitigation. They give even more reason to take action in controlling  $NH_3$ ,  $NO_x$  and VOC emissions to reduce their adverse impacts on health and ecosystems, while simultaneously tackling greenhouse gas emissions as the main cause of climate change.

## 4. Key Messages for Stakeholders & Societal Implications

ECLAIRE was structured around a set of key questions that was asked at the start of the project as a means to provide key messages for stakeholders. Initial answers to the questions were provided midway in the project, and have since been updated. Here we provide answers to these questions based on the most recent discussion of findings at the ECLAIRE final conference (Edinburgh, September 2015).

# Question 1: What are the expected impacts on ecosystems due to changing ozone and N-deposition under a range of climate change scenarios, taking into consideration the associated changes in atmospheric CO<sub>2</sub>, aerosol and acidification?

### Effects via atmospheric emission transport and deposition.

The main driver for future changes in N and  $O_3$  deposition will be changes in anthropogenic emissions, including those associated with adaptation to climate change, through changes in agricultural practice (management practice, crops selection), forestry (tree species selection), land-use and policy responses to climate change. However, the emissions are further modified through direct climate effects on the emission processes.

Climate change is expected to alter both the magnitude of primary emissions, especially from biogenic/agricultural sources (NH<sub>3</sub>, soil NO<sub>x</sub>, some BVOCs), as well as pollutant atmospheric lifetimes and resulting N deposition patterns. Results indicate that future climates are likely to increase NH<sub>3</sub> emissions strongly, along with increase in soil NO<sub>x</sub> in drying areas, which will propagate to increases in N deposition, especially close to source, and organic PM<sub>2.5</sub>.

A warmer climate is expected to increase BVOC emissions, while higher  $CO_2$  concentrations have a more complex effect.  $CO_2$  stimulates plant growth (enhancing BVOC emissions), but also dampens leaf-level emissions of some BVOC. In the case of isoprene the  $CO_2$  effect is expected to offset the temperature effect. There is insufficient evidence to conclude that  $CO_2$ trade-off will cancel a warming effect on monoterpene emissions. In addition, natural species adaptation and future human choices in agricultural and forest species in response to climate change may alter BVOC emissions significantly. The result is that net of climate change (directly and indirectly through land-use change) on tropospheric  $O_3$  remains less clear.

While precursor emissions will increase, the likely effect on inorganic  $PM_{2.5}$  concentrations is likely to be more complex. By contrast, climate change is expected to increase future N deposition through the warming effect, while anticipated changes in precipitation have a much smaller effect (only changing the location, but not the amount of deposition).

#### Effects via climate stress and extreme events

Effects of climate-related stress (drought, insect attack) and extreme events (fires, windfall, heavy rain) on emissions are likely to be significant but remain uncertain. For example, BVOC emission profiles have been found to be impacted by biotic stress (e.g. insect attack, drought stress), leading to profiles which result in more secondary organic aerosol formation. This means that plant biotic stress has impacts for human health, global dimming and further potential feedbacks on photosynthesis through increased aerosol loading. Climate change

impacts on biotic stress and its resulting feedbacks will need to be quantified better in future studies to judge whether it needs to be accounted for in mitigation policies.

### Interactions between air pollution and climate policies for nitrogen and methane

A significant off-set can be anticipated between changes in NO<sub>x</sub> and NH<sub>3</sub> emission changes considering anticipated climate change. While further reductions in NO<sub>x</sub> emissions can be expected over the  $21^{st}$  century (e.g. Gothenburg Protocol and NECD revision), climate induced increases in NH<sub>3</sub> emissions, combined with low take-up of available mitigation actions, will reduce the benefits of NO<sub>x</sub> controls for N deposition and PM<sub>2.5</sub> control. This result highlights the dual importance of a) applying available technical measures to reduce NH<sub>3</sub> emissions if adverse effects are to be avoided and b) ultimately incorporating climate sensitivity into official national NH<sub>3</sub> emissions inventories to properly account for this interaction.

Methane emission control is increasingly recognised as a win-win strategy whose control reduces climate change at the same time as reducing the production of  $O_3$ .

### Interactions on ecosystem responses to ozone and nitrogen

Plant productivity is generally increased by N and  $CO_2$ , and decreased by  $O_3$  and each of these effects may be altered under climate change.

Ozone pollution is likely to decrease Nitrogen Use Efficiency and increase N losses. Under elevated  $O_3$ , less N is used for growth, while plants are also less good at N resorption before litter-fall, so that more N is deposited to soils in leaf litter. The result is that  $O_3$  is likely to have knock-on effects by worsening nitrogen pollution, including and biodiversity changes, nitrate leaching and increased N<sub>2</sub>O emission.

Certain legumes are very ozone sensitive. This may lead to reduced N fixation in some ecosystems. Experimental evidence indicates that the differential sensitivity of species to ozone can lead to changes in community structure in developing grassland communities.

### Effects through nitrogen processes in forests

In N-limited forests, especially in boreal forests, N deposition enhances growth and carbon sequestration. Accumulated N deposition over time, however, tends to decrease C:N ratios in biomass, soil organic layer and to a lesser extent the soil mineral layer, and with a continuous elevated N input, the ecosystem may approach "N saturation".

In this stage, the N leaching will increase above background levels, associated with soil acidification in terms of elevated leaching of base cations or aluminium, causing relative nutrient deficiencies, which may be aggravated by a loss of mycorrhiza or root damage.

ECLAIRE has shown that positive impacts of N on growth occur below 15 kg N ha<sup>-1</sup> yr<sup>-1</sup>, but reverse between 15-25 kg N ha<sup>-1</sup>yr<sup>-1</sup>. One may consider 15 kg N ha<sup>-1</sup> yr<sup>-1</sup> a critical load for forest growth. At an N deposition below this load, there may still be adverse impacts on other forest ecosystem compartments, such as changes in ground vegetation and in mycorrhiza.

### Other ecosystem effects linking air pollution and climate

Increased temperatures are likely to increase species-richness, but also cause loss of coldtolerant species that may be important for conservation of biodiversity. Increased temperatures will increase N turnover, worsening effects of N on biodiversity and air- and water-quality in the short term, but potentially reducing accumulated N and so enhancing forest growth in N limited (especially boreal) forests.

Dryer soil and dryer air under climate change as well as elevated  $CO_2$  may reduce stomatal  $O_3$  uptake by vegetation and thus counteract adverse  $O_3$  effects. These changes also have other important effects on ecosystems which need to be considered, i.e. they are not generally positive. Chronic exposure to ozone can cause plants to be less tolerant of drought.

Longer growing seasons, higher temperatures (in cooler climates) and to some extent the climate change promotion of  $O_3$  formation will aggravate effects of  $O_3$ . In Northern Europe, an earlier start of the growing season may lead to an increasing overlap with the high  $O_3$  concentrations of the so-called  $O_3$  spring peak, possibly increasing environmental risks.

### Question 2: Which of these effects off-set and which aggravate each other, and how do the mitigation and adaptation measures recommended under climate change relate to those currently being recommended to meet air pollution effects targets?

### Interactions between pollution components

While N deposition generally increases rates of carbon uptake by vegetation, ozone reduces C uptake and storage in vegetation. The form of N deposition also affects the response. Overall,  $NH_3$  emission is associated with reduced N that contributes to C sequestration, but also has more adverse impacts on biodiversity than  $NO_y$  deposition. Conversely,  $NO_x$  emissions contribute to C sequestration, but also promote ozone formation that decreases C storage. Both components contribute to the cooling effect of aerosol, e.g. as ammonium nitrate, while deposition of both forms contribute to warming by increasing nitrous oxide ( $N_2O$ ) emissions. It should be noted that increases in C storage induced by N deposition are likely to be a transient benefit and will decrease in the future.

Although certain effects of pairs of environmental drivers, such as N,  $O_3$ ,  $CO_2$  and temperature, may be opposing, this cannot be extrapolated to say that effects by such pairs are cancelling each other out in general, since mechanisms of action are specific for the different environmental drivers. ECLAIRE has particularly shown that this is the case with the N and  $O_3$  interaction, where exposure to  $O_3$  can almost completely cancel the productivity benefit of N inputs in some ecosystems experiments.

### Interactions for forests

The enhanced forest growth and C sequestration since approximately 1950 up to 2005 is most likely mainly due to elevate N deposition. The implication of the ECLAIRE findings is that this would have been even larger in the absence of elevated ambient ozone concentrations. It seems that  $CO_2$  fertilization and temperature increase have so far played a comparatively minor role.

For the future, the expected forest growth is highly uncertain. When neglecting possible limitation by non-nitrogen nutrients (as is currently the case in nearly all earth system models), it seems likely that the expected large increase in  $CO_2$  and temperature will further enhance forest growth and C sequestration, especially in Central Europe. In southern Europe, more

limited water availability (drought stress) will most likely offset the growth enhancing effects of  $CO_2$  and temperature rise. For other parts of Europe, especially in N. Europe, these effects will most likely be compensated by limited N availability in view of expected decreased N deposition (N limiting the  $CO_2$  fertilization effect).

When accounting for the possible limitation by non-nitrogen nutrients, such as phosphate, calcium, magnesium and potassium, it is likely that no further increase in forest growth is to be expected because these nutrients will limiting growth, especially phosphorus.

### Other interactions to be considered

Several other changes can alter circulation of nitrogen in the environment and extent of ozone impacts. These include large scale land-use change, such as increased short-rotation forestry for biofuel production, which can change N deposition patterns as well as lead to increases or decreases in BVOC emission depending on species selection.

In addition, land-use changes that alter albedo of land can affect N and  $O_3$  effects. These include policies to avoid low albedo of farmland by reducing periods of bare soil and promoting high albedo in cities.

# Question 3. What are the relative effects of long-range global and continental atmospheric transport vs. regional & local transport on ecosystems in a changing climate?

Impacts of air pollution on European ecosystems occur over a range of spatial scales from the global scale ( $O_3$  background), though regional scale ( $O_3$  and N deposition) to local scale (N deposition and PM<sub>2.5</sub>, NH<sub>3</sub> exposure). In a changing climate, the spatial patterns of impacts are likely to change as a result of changing emissions, land use and atmospheric processes.

### Atmospheric transport changes for nitrogen compounds

Around 90-95% of impacts due to N deposition to European ecosystems are the result of European emissions. However, at a national level N deposition has contributions from both national emissions as well as emissions from neighbouring countries.

A warmer climate will most likely increase the relative contribution of  $NH_3$  to N deposition and thus increase near-source impacts relative to those at longer ranges. A warmer climate may also increase the evaporation of ammonium aerosol, leading to an increase in  $NH_3$  concentrations and may also affect the atmospheric lifetime of ammonia due to changes in compensation points.

Changes in precipitation patterns are likely to affect the spatial patterns of impacts as well. For example, reduced rainfall in southern Europe may increase the atmospheric lifetime of ammonium as a result of reduced wet deposition, leading to larger transport distances.

### Atmospheric transport changes for photochemical oxidants

Impacts of  $O_3$  in Europe are the result of pre-cursor emissions both from within Europe and worldwide. Summertime ozone concentrations in Europe are strongly influenced by European pre-cursor emissions whereas non-European pre-cursor emissions, of which methane is key, dominate the rest of the year.

A warmer climate could lead to increased water vapour, which would most likely decrease the  $O_3$  background, especially in summer, partially offsetting the effect of increasing non-European pre-cursor emissions. However, increasing temperatures could also decrease atmospheric sinks, such as the reaction with PAN, tending to increase  $O_3$  concentrations.

Reduced rainfall in southern Europe will increase the drought stress of vegetation, which would reduce  $O_3$  deposition in the region, thus mitigating ecosystem impacts to some extent but exacerbating the impacts to human health due to increased  $O_3$  concentrations.

As well as increasing temperatures and changing precipitation patterns, climate change is likely to alter global circulation patterns. Climate models predict an increase in atmospheric stagnation over Europe, which would exacerbate the impacts of  $O_3$ , especially those due to European precursor emissions.

# Question 4. What are the best metrics to assess O<sub>3</sub> and N impacts on plants and soils, when considering interactions with CO<sub>2</sub> and climate, and the different effects of wet vs dry deposition on physiological responses?

ECLAIRE has shown that, in contrast to concentration-based metrics, flux-based metrics that incorporate the modifying effects of climate, soil and plant factors on ozone uptake provide opportunities to incorporate the combined effects of pollutant interactions and climate change on plant response.

### Metrics related to nitrogen and its interaction with sulphur

Nitrogen deposition occurs in a number of different forms (i.e. wet and dry deposition,  $NH_x$  and  $NO_y$ ). The ECLAIRE experiments have demonstrated that direct effects, from atmospheric concentrations, are stronger when N is in the reduced form as ammonia. This points to the need for further development of effects metrics that distinguish the effects of  $NH_x$  and  $NO_y$ , dry/wet deposition on biodiversity. In contrast, there insufficient evidence to show that N effects mediated by soil processes depend on N form.

For N deposition effects on plant diversity, metrics should consider not only reduction in species diversity but also probabilities of the presence/absence of important species. This is important in the context of climate change, as plant species diversity may increase under a warming climate, while a simultaneous loss of key conservation species occurs.

For nitrogen (and S) a new biodiversity based indicator has been developed and mapped over Europe (Habitat Suitability Index). From this, preliminary thresholds for N (and S) deposition have been derived, and explored in integrated assessment (GAINS model). This indicator also depends on climate variables, and first tests of its climate sensitivity have been carried out. There is a need to further investigate the interpretation of these thresholds ("protection levels/loads"), especially for a non-expert audience given that the approach may appear to imply a different overall level of threat compared with previous approaches.

### Metrics to assess N and O<sub>3</sub> combinations

ECLAIRE has produced O<sub>3</sub> dose-response relationships for tree and crop species with novel response variables (e.g. net annual increment for forests; nitrogen use efficiency, protein and

starch yield, and grain mass yield for crops (wheat). Methods to incorporate the modifying effect of N on the sensitivity of these dose-response relationships have also been developed. These relationships can be used to: i) define scientifically determined 'no-effect' thresholds; ii) set policy relevant 'target' thresholds and iii) to quantify damage due to exceedance of the 'no-effect' threshold.

The interactions observed between N and  $O_3$  exposure in ECLAIRE are particularly significant. For example, the potential has been shown in field experiments for high  $O_3$  to negate the productivity benefits of N inputs. Such interactions point to the need to develop new metrics of N and  $O_3$  impacts that can take account of these interactions. For this purpose the development of process based models, such as  $DO_3SE$  at the plant scale and CLM and OCN on a global scale are providing a basis to start to assess the interactions.

There are clear interactive effects on plant species composition resulting from interactions between N deposition and  $O_3$  that occur over the short-term (1-2 years). However, there is no clear indication of whether these combined N and  $O_3$  effects will be positive or negative over the longer-term. Long-term monitoring of changes in plant species diversity with prevailing pollution concentrations and climate is essential to understand these dynamics better.

### Metrics to assess aerosol impacts on plant drought stress

ECLAIRE studies have indicated that reducing aerosol deposition to leaves may increase drought tolerance due to the removal of the wicking effect that can enhance water loss *via* stomata even when stomatal conductance is low. Experimental studies combined with monitoring of aerosol concentrations in a polluted part of central Europe have provided the basis to establish a first dose-response relationship between total hygroscopic aerosol concentrations. This approach provides the basis for model tests in DGVMs and also needs to be extended to consider the dose-response relationship for the overall stomatal response to drought.

### Question 5: What is the relative contribution of climate dependence in biogenic emissions and deposition vs. climate dependence of ecosystem thresholds and responses in determining the overall effect of climate change on air pollution impacts?

The findings of ECLAIRE indicate that climate change will occur through several mechanisms:

- Climate induced increases in emissions, especially of NH<sub>3</sub> from agriculture and NOx from agricultural and forest soils, but also some BVOCs, leading to increases in N deposition and a risk of higher O<sub>3</sub> concentrations.
- Climate induced changes in partitioning between aerosol and gas phases in the atmosphere leading to a relative increase in gas phase concentrations, such as NH<sub>3</sub> and nitric acid (HNO<sub>3</sub>), which may to some degree moderate expected increases in particulate matter concentrations under a future climate.
- Interactions with other atmospheric components, especially with parallel increases in CO<sub>2</sub> concentrations which are expected to moderate the increase in O<sub>3</sub> concentrations driven by the temperature effect on BVOC emissions.
- Changes in ecosystem vulnerability to a set concentration or flux of N or O<sub>3</sub> air pollution.

• Parallel changes in habitat suitability due to changing climate which combine with air pollution effects to further threaten sensitive plant communities.

With each of these interactions identified in ECLAIRE as being of significant importance it is hard to immediately generalise which of the factors is most important.

While the effects of temperature on biogenic and agricultural emissions are well established (NH<sub>3</sub>, some BVOCs, soil NO), effects of climate on ecosystem vulnerability will operate via alterations in drought stress, soil turn over processes and net photosynthesis. Drought may exacerbate some pollution effects such as limiting plant N uptake leading to larger N pollution losses in the environment and may be worsened under increasing background  $O_3$  exposure due to  $O_3$ -induced loss in stomatal control or due to aerosol deposition on leaf surfaces.

An ECLAIRE scenario was developed and analysed using the GAINS model. This compared the climate driven increase in  $NH_3$  emissions with the effect of climate change as an additional stressor on Habitat Suitability. While acknowledging uncertainties, for this analysis up to 2050, the effect of climate as an additional stressor to habitats was found to be even larger than the effect of climate on increasing  $NH_3$  emissions.

The key message, however, is that both of these type of factors are important. With some exceptions (like the  $CO_2$  effect) the changes mostly operate in the same direction: future climate change worsens the effects of air pollution on European ecosystems.

### Question 6: Which mitigation and/or adaptation measures are required to reduce the damage to "acceptable" levels to protect carbon stocks and ecosystem functioning? How do the costs associated with the emission abatement compare with the economic benefits of reduced damage?

### Mitigation and adaptation measures required

Experiments and analytical work in the project has further established evidence of the benefits of reducing nitrogen emissions. Lower  $NO_x$  emissions will reduce vegetation exposure to ground-level  $O_3$ , and thereby deliver positive benefits to forest growth and agricultural crops. While atmospheric deposition still fosters forest growth in N-limited regions of Europe, adverse conditions have been observed, especially in the long run, on biomass accumulation in regions more exposed to air pollution.

Balancing the  $O_3$  damage and biodiversity loss from N against its contribution to possible increases in C stocks and productivity remains a complex task, especially with respect to economic considerations. ECLAIRE has highlighted the wider issues which will likely need to be considered as context along with an ecosystem services based analysis, to reflect on the problem comprehensively.

Precursor emissions that affect background  $O_3$  on the hemispheric scale are proving to be important in determining exposure of vegetation to ground level  $O_3$  (especially methane). Further reductions of ozone fluxes in Europe require tackling precursor emissions at the hemispheric scale (especially of methane).

Ammonia and  $NO_x$  reduction is also beneficial to reduce PM exposure and human health effects Cost-effective health driven air pollution policy will also reduce excess nitrogen on nature. For ammonia low cost measures are widely available, especially for large farms.

### Costs compared with the benefits of emission abatement

Benefits of a scenario implementing maximum technical reductions in the EU for crops, timber production and carbon sequestration are €1.8 billion. Less excess nitrogen deposition will also contribute to the achievement of existing biodiversity commitments.

Support provided by ECLAIRE to the Gothenburg Protocol and NECD revision process has highlighted that mitigation measures for  $NO_x$  are becoming increasingly expensive, while many low-cost mitigation options for  $NH_3$  have not yet been adopted in many countries. This is illustrated in **Figure 29**, which shows the benefit/cost ratio for further air pollution mitigation beyond existing commitments for 2020, including estimates of health and ecosystem costs vs the cost of mitigation actions. The current position as illustrated by this graphic suggests that a further 1100 kt  $NH_3$ -N mitigation is cost optimal, but only a further 300 kt  $NO_x$ -N mitigation.



**Fig. 29:** Comparing benefit/cost ratios of reducing reduced nitrogen with that of reducing oxidized nitrogen emissions (Van Grinsven et al., 2013).

Health driven air pollution policy will also reduce excess nitrogen on nature by ~44%. An illustrative ECLAIRE scenario that reduces excess deposition with 2% more will cost  $\leq$ 23 mln. The benefits of such an additional reduction will be 50-1000% higher, depending on the methodology for biodiversity valuation.

### Wider approaches to air pollution mitigation strategies

Additional nitrogen reduction is needed to keep the risks for biodiversity constant in a changing climate. New technologies and structural changes in production and consumption will be needed to increase the scope for further reductions in excess nitrogen deposition and ozone fluxes. Increased nitrogen use efficiency will lead to cost savings in food production and consumption on a longer time scale (Sutton et al., 2013, Sutton and Bleeker, 2013).

The issue of food consumption is closely linked to the nitrogen cycle given the major role of nitrogen in food and feed production and in livestock rearing. A special report facilitated through ECLAIRE in partnership with the UNECE Task Force on Reactive Nitrogen, "Nitrogen

on the Table" found that halving consumption of meat and dairy products across Europe would lead to around a 40% reduction in Nitrogen pollution, while liberating large areas of agricultural land for other uses (e.g. bioenergy production). Overall the nitrogen use efficiency of the European food system was doubled under this scenario (Westhoek et al., 2015).

# Question 7: How can effective and cost-efficient policies on emission abatement be devised in the future?

The results from the ECLAIRE project continue to demonstrate that an integrated approach to addressing the scientific questions is necessary to develop an integrated policy perspective. This integration then allows the selection of win-win scenarios or informs prioritisation needs, which leads to more effective policies. It turns out that the most effective way forward is to reduce emissions of  $NH_3$  in Europe to halt the loss of biodiversity, and of  $CH_4$  at the hemispheric scale to reduce ozone damage. Specific actions are as follows:

- Reducing nitrogen deposition has benefits for both ecosystems biodiversity and human health. This allows for cost sharing during implementation of measures, which increases their overall cost-effectiveness. The first cost-benefit analyses for ecosystems from ECLAIRE can therefore support the development of integrated cost-effective policy.
- While N deposition enhances net primary production of ecosystems in the short term in N limited areas, excess N may have negative effects on biomass growth in the long run. This points to further benefits in reducing nitrogen emissions in Europe
- Decreasing NH<sub>3</sub> has both health and ecosystem benefits with low cost measures available.
- Action on methane will have benefits for both air pollution and climate but will require hemispheric integration of the relevant policies to maximise effectiveness.
- Monitoring is an essential part of the process, from establishing current trends through to gauging the impact of policy measures.
- Adopting a win-win approach may require broader top-down policymaking strategies, which make the consideration of more than one pollutant or sector more achievable. At the least a more integrated consideration of the range of issues is needed.
- Policy is most effective when it has the support of the general public, therefore increasing efforts to communicate clear messages on effects and solutions is essential.
- The multiple effects of nitrogen pollution across the nitrogen cycle link air and water pollution, climate change and biodiversity. A joined-up nitrogen strategy would therefore have benefits in overcoming barriers-to-change, highlighting win-win for businesses and the environment. The ECLAIRE community is stimulating this activity through its leadership of the International Nitrogen Management System (INMS) in cooperation with the UN Environment Programme (UNEP) and the International Nitrogen Initiative (INI).
- Behavioural changes offer a very important part of the suite of available solutions, to reduce air pollution impacts on ecosystems. Highlighting effects on cherished species, the cobenefits of improved diet for health and engaging the public in data gathering through citizen science activities may aid in the process.

In addition to the underpinning science, ECLAIRE has been extremely active in providing support for European policy development. Key outcomes include support to the EU policy review (e.g. Fowler et al., 2013; Brunekreef et al., 2015), guidance on pollution mitigation and costs (Bittman et al., 2014; Reis et al., 2015; UNECE, 2015) and examination of the pollution and land use relationships for future food choice scenarios (Westhoek et al., 2015).

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Part 2: Plan for the Use and Dissemination of Foreground

2.1. Section A1: List of all scientific publications relating to the foreground of the project.

2.2. Section A2: List of all dissemination activities

## 2.3. Section B1: Exploitable foreground information (patents etc)

Not applicable

## 2.4. Section B2: Overview table with exploitable foreground

TYPE OF EXPLOITABLE FOREGROUND	DESCRIPTION OF EXPLOITABLE FOREGROUND	CONFIDENTIAL	FORESEEN EMBARGO DATE DD/MM/YYYY	EXPLOITABLE PRODUCT(S) OR MEASURE(S)	SECTOR(S) OF APPLICATION	TIMETABLE FOR COMMERCIAL USE OR ANY OTHER USE	PATENTS OR OTHER IPR EXPLOITATION (LICENCES)	OWNER AND OTHER BENEFICIARY(S) INVOLVED
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Datasets on trace gas and aerosol fluxes from the 10- site ELCAIRE network	No	N/A	Datasets uploaded to the ECLAIRE database	Science community	N/A	N/A	NERC and ECLAIRE partners
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Datasets on ecosystem responses to nitrogen, ozone and aerosol pollution	No	N/A	Datasets uploaded to the ECLAIRE database	Science Community	N/A	N/A	NERC and ECLAIRE partners
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Updated mathematical models of trace gas and aerosol fluxes, atmospheric chemistry and transport, ecosystem responses and cost- benefit analysis	No	N/A	Models maintained by relevant project partners	Science Community	N/A	N/A	NERC and ECLAIRE partners
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	New model ECLAIRE Surface eXchange (ESX) of trace gas and aerosol surface interactions	No	N/A	New model maintained by MET.NO and FMI on behalf of the consortium	Science Community	N/A	N/A	MET.NO, FMI, NERC, UoY, INRA and other ECLAIRE partners

SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	New fundamental understanding and expertise on N-O3 interactions	No	During 2016 when key paper published (date not yet set).	Data for high profile and other key publiccations	Science community	N/A	N/A	NERC and ECLAIRE partners
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	New fundamental understanding on aerosol – drought interactions for plants	No	During 2016 when key paper published (date not yet set).	Data for high profile and other key publiccations	Science community	N/A	N/A	NERC, UBONN and ECLAIRE partners.
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Data contained within the 'N on the Table Report'.	No	12 <sup>th</sup> January 2016, Report launch planned for European Parliament	Environmental consequence of dietary change scenarios	General Public, Food industry	N/A	N/A	NERC, JRC, PBL and other ECLAIRE partners
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Expertise and relationships on agriculture-climate relationships and policy relevance	No	9 <sup>th</sup> December 2015, Presentation of evidence to UK parliament EFRA Select Committee	Potential and cost effectiveness of agricultural ammonia mitigation	General Public, Food industry	N/A	N/A	NERC and ECLAIRE partners.
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Expertise and relationships between air pollution effects on ecosystems, damage costing and implication for EU Air Quality Policy	No	N/A Interventions through 2016 to support National Emissions Ceilings Directive with COMM, EU Parliament and Member States	Cost effectiveness of mitigation options under climate change and considering the ecosystem interactions.	General Public, Food industry	N/A	N/A	NERC

SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Expertise on nitrogen cycle in relation to air pollution and wider interactions in policy context.	NO	N/A Ongoing. Application to development of International Nitrogen Management System (INMS) with UNEP	Expertise and opportunities for joined up approach on nitrogen management to help overcome the barriers to change.	Policy Makers, General Public, Industry.	N/A	N/A	NERC and ECLAIRE partners.
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Expertise and models on air pollution- climate interactions for ecosystems	NO	N/A Ongoing. Application to future strategy of the UNECE Conventon on Long-range Transboundary Air Polluiton	Expertise and analysis inputs to the different LRTAP groups, including Executive Body, Working Group on Strategies and Review, Working Group on Effects, EMEP etc.	Policy Makers, General Public, Industry.	N/A	N/A	NERC and ECLAIRE partners.
SOCIAL INNOVATION & EXPLOITATION OF RESULTS THROUGH EU POLICIES	Expertise on inter- media nitrogen pollution understanding, control options and co-benefits.	NO	Workshop to be announced during LRTAP Executive Body (Dec 2015) and held during May 2016. (Funding is available from DG Env).	Post project workshop funded by DG Env towards a joined-up European guidance on good N management for air, water, climate co- benefits.	Policy Makers, General Industry.	N/A	N/A	NERC and ECLAIRE partners.

### Datasets on trace gas and aerosol fluxes from the 10-site ELCAIRE network

Flux datasets collected as part of ECLAIRE will provide a key resource following the end of the project for further testing and development of air pollution transport and surface exchange models. The dataset are archived in the ECLAIRE database. A key user group will be the European Monitoring and Evaluation Programme (EMEP) under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP)

### Datasets on ecosystem responses to nitrogen, ozone and aerosol pollution

Datasets on the ecological impacts of air pollution collected as part of ECLAIRE will provide a key resource following the end of the project for further testing and development air pollution effects metrics. The dataset are archived in the ECLAIRE database. A key user group will be the Working Group on Effects (WGE) and its component Task Forces under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP)

# Updated mathematical models of trace gas and aerosol fluxes, atmospheric chemistry and transport, ecosystem responses and cost-benefit analysis

Atmospheric transport models developed and improved as part of ECLAIRE will provide a key resource following the end of the project for the European Monitoring and Evaluation Programme (EMEP) under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP).

# New model ECLAIRE Surface eXchange (ESX) of trace gas and aerosol surface interactions

Under ECLAIRE a completely new surface-atmosphere interactions model has been developed called ESX, which links air pollution interactions with climate variables using a process based description. This will provide a key resource for further development as part of the European Monitoring and Evaluation Programme (EMEP) under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP).

### New fundamental understanding and expertise on N-O<sub>3</sub> interactions

The advances in ECLAIRE provide a fundamental step-change in our understanding of N and O<sub>3</sub> interactions when considering the effects of air pollution on ecosystems. The new findings highlight the knock-on consequences of O<sub>3</sub> pollution for other forms of nitrogen pollution, worsening water quality and greenhouse gas emissions through nitrates and nitrous oxide. These results will be exploited by development of

high profile publications into this topic which will be subject to press embargo before publication.

### New fundamental understanding on aerosol – drought interactions for plants

ECLAIRE has made fundamental advances in understanding the interactions between aerosol and drought effects on plants which are highly relevant under climate change. Specifically, the combination of detailed experimental studies with ambient monitoring has allowed a first dose response concept to be established between hygroscopic aerosol loading (part of the PM fraction) and effects on stomatal conductance. This will be exploited after ECLAIRE by testing in Dynamic Global Vegetation Models (DGVMs), followed by key publications which may also need to be embargoed prior to publication.

### Data contained within the 'N on the Table Report'.

The purpose of the data contained within the report is to highlight the potential benefits from reduction of the consumption of animal protein, described by a number of scenarios. The benefits described are reduction in nitrogen pollution to air and water and the potential impacts of dietary change on land-use. This information could be exploited to inform the future development of water and air pollution policy and engage the public in understanding the impact of their lifestyle choices. The Key Messages were launched in 2014 with substantial press reaction (including The Times, Press Association, BBC etc). The full report will be launched at the European Parliament in (provisionally 12) January 2015 and will be embargoed until 13 January.

### Expertise and relationships on agriculture-climate relationships and policy relevance

The expertise developed by ECLAIRE is proving of interest to many policy stakeholders. This has now been followed by an invitation to give evidence to the UK House of Commons Select Committee on Environment Food and Rural Affairs (9 December 2015). Written evidence has already been submitted and will be available in due course at: <u>http://www.parliament.uk/business/committees/committees-a-z/commons-select/environment-food-and-rural-affairs-committee/news-parliament-2015/air-quality-evidence-15-16/</u>

## Expertise and relationships between air pollution effects on ecosystems, damage costing and implication for EU Air Quality Policy

The expertise developed in ECLAIRE has been called upon on several instances during the project to provide support for EU Air Quality Policy, including ECLAIRE experts speaking at several hearings of at the European Parliament and Brussels Green Week. As the EU Air Quality Package is negotiated with Member States following the vote of MEPs on the Commission Proposal in October 2015, it is expected that ECLAIRE experts and data will continue to be called on during 2016 to support the European Commission and Member States.

# Expertise on nitrogen cycle in relation to air pollution and wider interactions in policy context.

ECLAIRE has contributed to developing a wider perspective in relation to air pollution and other pollution threats (water pollution, climate change, human health etc) mediated through human alteration of the nitrogen cycle. The expertise and findings are feeding into development of a new science support process for international nitrogen policy, the "International Nitrogen Management System" (INMS). ECLAIRE outcomes will continue to feed into this process in 2016 and beyond, working in close engagement with the United Nations Environment Programme (UNEP) and the International Nitrogen Initiative (INI).

### Expertise and models on air pollution-climate interactions for ecosystems

ECLAIRE was specifically designed to provide science support to the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP). As such the ECLAIRE team is closely embedded into the LRTAP bodies, from the Executive Body (EB) and the Working Group and Strategies and Review (WGSR) to the EMEP Steering Body and the Working Group on Effects (WGE). The substantial advance in understanding from ECLAIRE will feed into the LRTAP process through 2016 and beyond.

### Expertise on inter-media nitrogen pollution understanding, control options and cobenefits.

Following the successful leadership by the ECLAIRE team of the UNECE "Framework Code for Good Agricultural Practice for Reducing Ammonia Emissions" (http://www.unece.org/index.php?id=41358&L=0) the European Commission, DG Environment, has been successful in obtaining additional funds to hold a workshop on Developing Guidance for Good Nitrogen Management to Achieve Co-benefits for Air, Water, Biodiversity and Climate. This workshop will be announced during the Executive Body of the LRTAP Convention and is planned for May 2016. Part 3: Report on Wider Societal implications of ECLAIRE

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