Project Number 282910

ÉCLAIRE

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

Seventh Framework Programme

Theme: Environment

D2.4 Definition and improved parameterization of fluxes of BVOC

Due date of deliverable: **01/04/2015** Actual submission date: **30/09/2015**

Start Date of Project: **01/10/2011** Duration: **42 months**

Organisation name of lead contractor for this deliverable: **Juelich**

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

1. Executive Summary:

New parametrizations were achieved for:

- *de-novo* emissions as function of the volumetric water content
- stress induced emissions relative to constitutive monoterpene emissions for regional models
- basal emission factors of isoprene emissions from oak species
- bidirectional flux of isoprene degradation products

2. Objectives:

Definition and improved parameterization of fluxes of BVOC under new environmental constraints and in relation to pollutants and endogenous induced emissions of NO and reactive oxygen species

3. Activities:

3.1 Parametrization of *de-novo* emissions as a function of the volumetric water content

Impacts of soil moisture on *de-novo* monoterpene (MT) emissions from Holm oak, European beech, Scots pine, and Norway spruce were studied in laboratory experiments. The volumetric water content of the soil, θ, was used as the reference quantity to parameterize the dependency of MT emissions on soil moisture and to characterize the severity of the drought.

3.2 Parameterization of stress induced emissions relative to constitutive monoterpene emissions

We developed a very simple parametrization to implement stress induced emissions in regional models for first estimates of their impact in atmospheric chemistry (here on SOA formation). Emission ratios of SIE/MT as determined in Jülich Plant Atmosphere Chamber served as basis (Mentel et al, 2013, Kleist et al. 2012). The parametrization was implemented in the EMEP MSC-W model (Simpson et al., 2012; Bergström et al., 2012).

3.3 Derivation of basal emission factors of isoprene emissions from oak species

Recognising that >60% of Europe's biogenic isoprene emissions are thought to be emitted by oak trees, a total of five datasets of canopy-scale isoprene flux measurements above forests dominated by oaks (from ECLAIRE flux sites and beyond) were analysed to almost double the database on which European emission calculations are based. This analysis focussed on the sensitivity of the derived basal emission factors on the modelling framework used to link leaf level emission factors to canopy scale fluxes and provided an uncertainty analysis of the process.

3.4 Definition of the bidirectional flux of isoprene degradation products

Methyl vinyl ketone (MVK) and methacrolein (MAC) are key oxidation products (iox) of isoprene, the most abundant volatile organic compound (VOC) emitted by vascular plants in the atmosphere. Increasing attention has been dedicated to iox, as they are involved in the photochemical cycles ultimately leading to ozone (O_3) and particle formation. However, the capacity of plants to exchange iox under low and realistic ambient concentrations of iox needs to be assessed. We hypothesized that a foliar uptake of iox exists even under realistic concentrations of iox. We tested the capacity of iox exchange in trees constitutively emitting isoprene (*Populus nigra*) or monoterpenes (*Quercus ilex*), or that do not emit isoprenoids (*Paulownia imperialis*). Laboratory experiments were carried out at the leaf level using enclosures under controlled environmental factors and manipulating isoprene and reactive oxygen species (ROS) production by using the isoprene specific inhibitor fosmidomycin, acute O_3 exposure (300 ppbv for 4 h), and dark conditions. We also tested whether stress conditions inducing accumulation of ROS significantly enhance iox formation in the leaf, and their emission.

4. Results:

4.1 Parametrization of *de-novo* emissions as function of the volumetric water content

When θ dropped from 0.4 $m^3 \cdot m^{-3}$ to ~ 0.2 $m^3 \cdot m^{-3}$ slight increases of *de-novo* MT emissions were observed but with further progressing drought the emissions decreased to almost zero (Fig. 1).

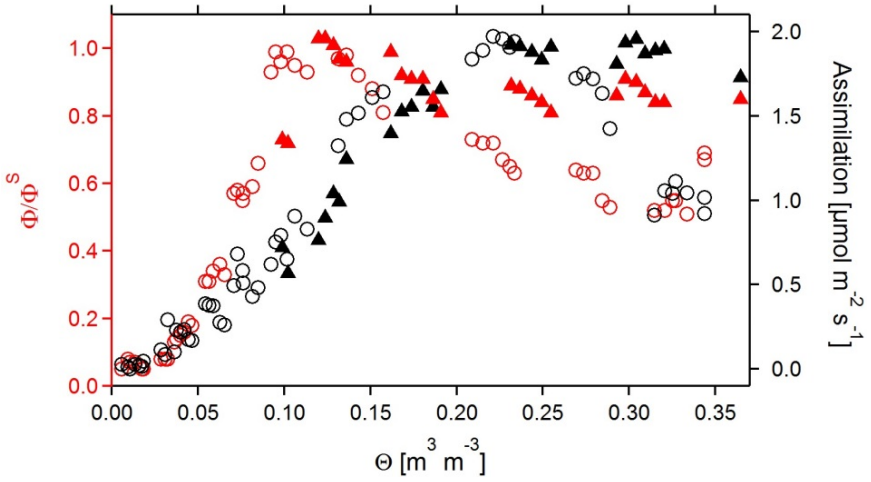

Fig. 1: Normalised sabinene emissions from beech (red symbols, left scale) and rates of net photosynthesis (black symbols, right scale, multiplied by -1) in dependence on θ. Closed triangles represent data taken during a first drought period that was stopped when θ had fallen to 0.1 $m^3 \cdot m^{-3}$, open circles represent data taken during the following period of recovery until the end of the severe drought at day 17. Only data taken at a chamber temperature of 23 °C and a PAR of 440 µmol m^{-2} s^{-1} are considered.

In most cases the increases of MT emissions observed under conditions of mild drought were explainable by increases of leaf temperature due to lowered transpirational cooling. When θ fell below certain thresholds (termed as θ_1), MT emissions decreased simultaneously with θ and the relationship between θ and MT emissions was approximately linear. In order to parameterize of *de-novo* MT emissions, three regimes of θ were defined:

$$\gamma_{SM} = 1 \qquad \theta > \theta_1$$

$$\gamma_{SM} = \frac{\theta - \theta_0}{\Delta\theta_1} \qquad \theta_0 < \theta < \theta_1$$

$$\gamma_{SM} = 0 \qquad \theta < \theta_0$$

where γ_{SM} describes variations of emissions due to soil moisture θ. Above the threshold θ_1, emissions are not directly affected by soil moisture, i.e. γ_{SM} = 1. Below θ_1, emissions linearly decrease with decreasing θ until θ_0, $\gamma_{SM} = (\theta-\theta_0)/\Delta\theta_1$, $\Delta\theta_1 = \theta_1-\theta_0$. Below θ_0, the extrapolated emissions became zero, γ_{SM} = 0. The thresholds of θ (0.044–0.19 $m^3 \cdot m^{-3}$) were determined as well as other parameters required to describe the soil moisture dependence of *de-novo* MT emissions for application in the Model of Emissions of Gases and Aerosols from Nature, MEGAN. A factorial approach was found appropriate to describe the impacts of θ, temperature, and light. The use of θ as reference quantity in a factorial approach was tenable in predicting constitutive *de-novo* MT emissions when θ changed on a time scale of days.

4.2 Parametrization of stress induced emissions relative to constitutive monoterpene emissions

The parametrization requires as an input the constitutive MT emissions used by the model (here standard EMEP emissions for monoterpenes, Simpson et al. 2012). Depending on the stress scenario emission ratios for SIE/MT are applied based on experimental data from JPAC (Mentel et al., 2013) to set the SIE in the model. The SIE considered here (SQT and MeSA) are of *de-novo* type (Kleist et al., 2012), i.e. they are emitted in connection with biosynthetic production. The emission ratios were determined in JPAC under steady state conditions as described above. The direct use of JPAC derived data for application or extrapolation to ambient conditions has been confirmed in earlier studies for particle formation rates and SOA mass yields for boreal tree species

(Mentel et al., 2009), the chemical composition of the resulting SOA (Kiendler-Scharr et al., 2009b), interaction of isoprene and MT emissions (Kiendler-Scharr et al., 2009a), and the distribution of highly oxidized aerosol precursors (Ehn et al., 2012).
From the observations by Mentel et al. (2013), we constructed three biotic stress scenarios.
Case 1 treats aphid infestation with enhanced SQT emissions with SQT/MT = 2.4 (mass based ratios).
Case 2 covers aphid infestations which caused enhanced emissions of SQT (SQT/MT = 4.9) and triggered MeSA emissions via the shikimate pathway (MeSA/MT = 3.75).
Case 1 and Case 2 were assumed to be typical for all trees of boreal and central European forests. As the SIE emissions studied here are of *de-novo* type, they were only switched on during daytime.
Since the observed emission factors only consider infested trees, the fraction of infested trees had to be estimated. We based the (rough) estimate on regular surveys of the European forests. ICP Forests (the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests operating under the UNECE Convention on Long-range Transboundary Air Pollution) provides annual executive reports on the conditions of the forests in Europe (http://www.icp-forests.org/); they also publish reports of the national member forest agencies. Fischer et al. (2012) provide details of tree crown damage and defoliation in many European countries; they report that a fifth of all trees are rated as damaged and that “defoliation represents a valuable early warning system for the response of forest ecosystems to different stress factors”. They also report that insects are the most frequent cause of damage to trees in Europe.

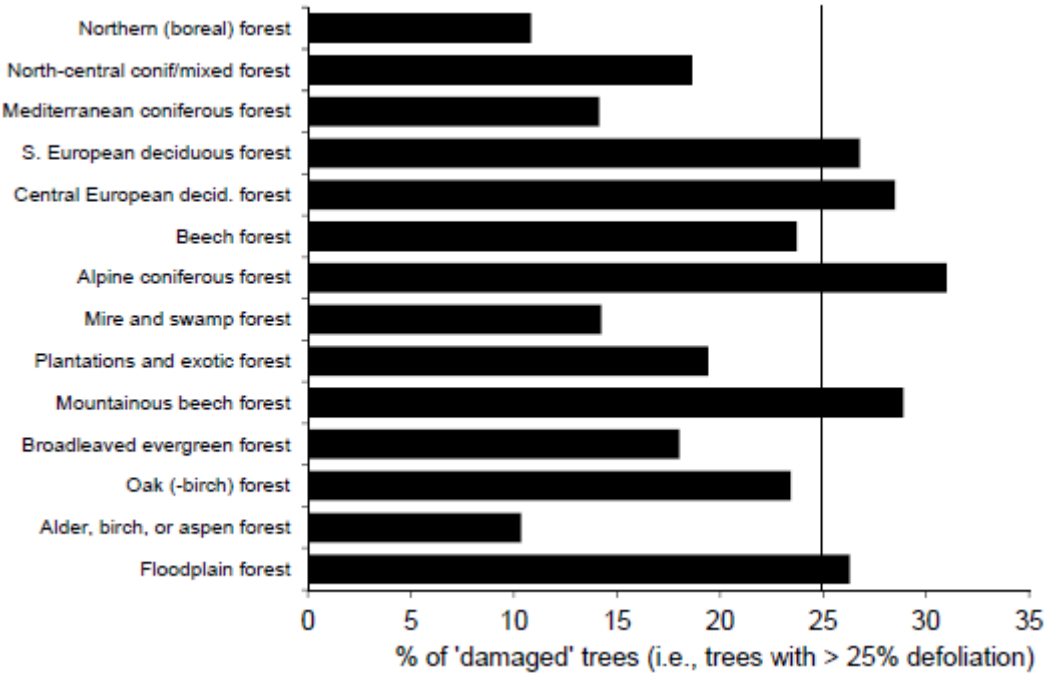

Fig. 2: Fraction (%) of damaged trees (>25% defoliation) in different European forest ecosystems. The top two bars refer to the main forest types investigated in the present study. (Figure adapted from ICP Forests, Fischer et al., 2012, used with permission.)

Relatively large fractions of the northern and central European trees show a significant degree of defoliation (>25%, rated as damaged by Fischer et al., 2012); the fractions of damaged trees are 11% for northern boreal forests, 19% for north–central coniferous/mixed forests (e.g. Germany and southern Sweden), 28% for central European deciduous forests, and 24% for beech forests, as can be seen in the top two bars and 4th to 5th bar of Fig. 2. We suggest that the reported defoliation damage can be used as a measure of stress effects. The applicability of our assumption is supported by the large European forest survey 2010 (ICP Forests, 2011), which found that insects are the greatest cause of damage to trees; 17% of all investigated trees were damaged by insects (corresponding to 27% of the total number of damaged trees). For comparison, about 10% of the trees were damaged by “fungi” and ca 9% by abiotic factors (e.g. drought and frost) (for more details about different damaging agents see Fig. 2–5, in ICP Forests, 2011). Insect infestations are well distributed over northern and central European forests as shown by Fischer et al. (2012) (see their Fig. 2–3). For the

boreal forests, the Finnish forest damage report from Merilä et al. (2007) stated that about 10–12% of the pines showed a significant degree of defoliation (>25%). Similarly to the European situation, about the same order of trees (about 10%) showed damage caused by insects and fungi, with in general more being caused by fungi. By combining these two types of forest observations, we conclude for our cases that the fraction of trees with significant defoliation (>25%) may serve as a first order approach to assess the fraction of stressed trees in the current situation. Consequently, we adopt these numbers and assume that the fraction of currently stress-affected trees is 10% for latitudes greater than 60° N and 20% between 45°N and 60°N. This constituted our base case for impact of contemporary SIE (Case 1 and Case 2). As an illustration Fig. 3 shows the base case and the effect of SIE on SOA formation.

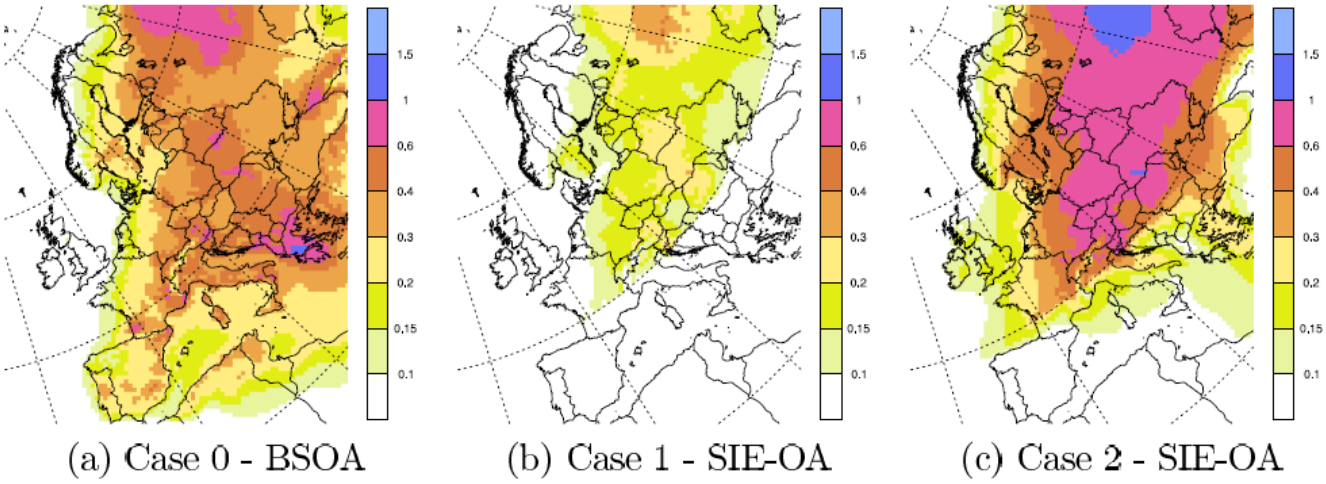

Fig. 3: Model calculated 6-month mean (Apr–Sep) concentrations of BSOA and biotic stress-induced OA (SIE-OA); (a) BSOA in Case 0 (reference case without explicit stress-induced emissions), (b) SIE-OA in Case 1 (biotic stress with sesquiterpene (SQT) emissions), (c) SIE-OA in Case 2 (biotic stress with emissions of SQT and methyl salicylate). Unit: μgm^{-3}.

4.3 Isoprene emission factors from oak species

Keenan et al., (2009) recently developed a process based inventory of isoprenoid emissions from European forests by first compiling a state-of-the-art database of canopy-scale isoprene basal emission factors (30 °C and 1000 µmol PAR s^{-1}) scaled from leaf-level measurements and subsequently using these values to initialize regional-scale isoprene emission models. These analyses highlighted the importance of Oak species to the European isoprene emissions budget with *Quercus robur*, *Quercus pubescens* and *Quercus petrea* thought to account for 63% of the total. However, this figure is highly uncertain as the basal emission factors upon which the model runs are based relate to values obtained from a handful of leaf-level studies which may not be particularly representative given the very large variability in emission rates between individual leaves.

Emission factors can also be derived from top-down micrometeorological flux measurements which are integrated over large areas (the flux footprint) and are therefore thought to be much more representative than leaf-level emission factors which must be up-scaled to the canopy level. Using isoprene measurements from two ECLAIRE core flux sites (Bosco Fontana & Ispra, both Italy) as well as three existing datasets from further sites (Alice Holt, UK; Castelporziano, Italy; Observatoire de Haute Provence (O3HP), France) within Europe, we calculated new canopy-scale basal emission factors for use in BVOC emission models. Each of the sites used in this study are described in Table 1.

Table 1: Description of the species composition, analytical methods used and canopy description for each of the five measurement sites used in this study.

	Alice Holt, UK (AH)	Bosco Fontana, Italy (BF)	Castelporziano, Italy (CP)	Ispra Forest, Italy (IF)	Observatoire de Haute Provence, France (O3HP)
Species composition	• *Quercus robur* (Pedunculate Oak, 90%) • *Quercus petraea* (Sessile Oak) • *Fraxinus* (Ash, 10%)	• *Quercus robur* (Pedunculate Oak) • *Quercus cerris* (Turkey Oak) • *Quercus rubra* (Northern Red Oak) • *Carpinus betulus* (Hornbeam)	• *Pinus pinea* (stone pine) • *Quercus ilex* (holm oak) • *Quercus suber* (cork oak).	• *Quercus robur* (Pedunculate Oak, 80%) • *Alnus glutinosa* (Black Alder, 10%) • *Popolus alba* (White poplar, 5%) • *Carpinus betulus* (Hornbeam, 5%)	• Quercus pubescens (Downy Oak, 75%) • Acer monspessulanum (Montpellier Maple, 25%)
Method, Analyser	vDEC, PTR-MS	vDEC, PTR-MS	vDEC, PTR-MS	EC, FIS	vDEC, PTR-MS
LAI [m^2/m^2]	4.8	5.5	4.6	4.4	2.4
Z [m]	28.5	32	35	37	10
h [m]	20.5	28	25	26	5

Basal emission rates for isoprene were calculated at each site by normalising the measured fluxes to a set of standard conditions (30 °C and 1500 μmol s^{-1} photosynthetically active radiation (PAR)) using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al. 2006; Guenther et al. 2012). MEGAN predicts isoprene emission rates at the canopy-scale by multiplying a known, species or ecosystem specific emission factor, ε, by an activity factor γ, which accounts for emission response to environmental factors such as light (γ_P), leaf temperature (γ_T), soil moisture (γ_{SM}), leaf age (γ_A), leaf area index and CO_2 inhibition (γ_C)

$$F_{iso} = \varepsilon \times \gamma$$

The MEGAN model was initialised at each of the five sites using the observed meteorological parameters and run in a mode that also took into account the influence of the previous 24 and 240 hours of temperature and light. In each model run the ambient air temperature was first converted to leaf temperature using an energy balance approach but the effects of CO_2 enrichment and soil moisture were not included. At each site a time series of the activity factor γ was generated from MEGAN by setting the isoprene emission factor within the model to unity. Measured fluxes were then plotted against the γ values and the new canopy-scale emission factor was taken as the slope of the regression with an uncertainty of three sigma. Unless measuring above a monoculture, this value can be thought of as an ecosystem-level emission factor as it reflects the forest canopy as a whole which includes multiple tree species. In order to derive emission factors specific to oak, the ecosystem-level emission factors were scaled based on the known species composition at each site to give an emission factor representative of 1 m^2 of pure oak forest. This approach assumes that isoprene emissions at each site were solely attributable to oak species. Based on the species present this appears justified at four of the five sites but at the Ispra forest site isoprene emissions may also have come from *Popolus alba*, a known isoprene emitting species. However, *Popolus alba* only accounted for 5% of the species present and therefore its contribution was thought to be relatively minor.

Fig. 4 shows the isoprene emission factors derived from micrometeorological flux measurements made at the two ECLAIRE flux sites and a further three European forest sites. The reported emission factors vary considerably which reflects both the mix of oak species present at each site and the structure of the forest canopy itself which determines the proportion sunlit and shaded leafs and ultimately the emission rate. For example, the isoprene emission factors for the two sites dominated by *Quercus robur* (e.g. Bosco Fontana (90% *Quercus rubur*) and Ispra) are broadly similar but the slightly

larger emission potentials at the Ispra site likely reflect the lower LAI and therefore higher proportion of sunlit leaves. By contrast the emission factor derived for the other Italian forest, Castelporziano are very low. This relates to the presence of *Quercus ilex* and *Quercus suber*, two oak species known to emit large quantities of monoterpenes but very little isoprene. Comparisons of emission factors between the remaining sites are difficult to make due to the differing species composition.

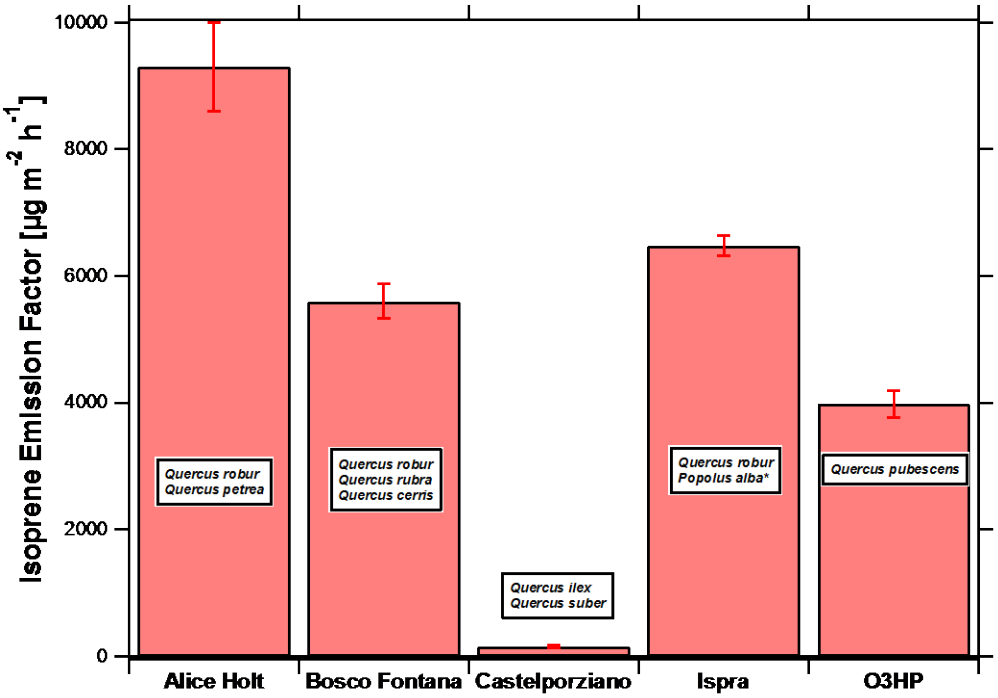

Fig. 4: Standardised (30 °C and 1500 µmol PAR s^{-1}) isoprene emission factors from European Oak forests based upon canopy-scale eddy covariance flux measurements. Emission rates are reported per square meter of Oak forest and thus have been corrected on the basis of the species composition reported at each site. *A small percentage of the Ispra forest site was comprised of *Popolus alba* (5%) which is also a known isoprene emitter.

The new isoprene emission factors were compared to the leaf-level derived emission factors presented by Kennan et al. (2009). Such a comparison is not straight forward due to the different algorithms used to normalize both leaf-level and canopy-scale flux data to standard conditions. The leaf-level measurements used by Keenan et al. (2009) were normalized to 30 °C and 1000 µmol s^{-1} PAR and subsequently scaled to the canopy level using a projected leaf dry mass per unit area. By contrast, our canopy-scale emission factors calculated using the MEGAN algorithms use a value of 1500 µmol s^{-1} PAR as standard conditions and assume a canopy LAI of 5. Therefore, in order to make a direct comparison between emission factors it was first necessary to re-normalise the canopy-scale emission factors to the same set of standard conditions as used by Keenan et al. (2009).
Table 2 shows the adjusted oak emission factors which reveal the values of Keenan et al. (2009) to be within the range of those calculated for *Quercus robur* but to slightly overestimate the emission potentials of *Quercus pubecens* and *Quercus rubra*.
It should be noted that the emission factors derived from flux measurements are likely to underestimate the true emission potential of the forest. This is because the micrometeorological flux measurements upon which they are based represent the net exchange occurring above the canopy which is a balance between isoprene emission and dry deposition.

Table 2: Summary of canopy-scale isoprene emission factors. For comparison with the leaf-level emission factors reported by Keenan et al., (2009) the presented emission factors have been normalised to 1000 µmol s^{-1} of PAR and reflect the site specific LAI values listed in Table 1.

	Keenan et al. (2009)	Alice Holt	Bosco Fontana	Castelporziano	Ispra	O3HP
Q. robur	6819	8271±622	4985±241	-	5764±137	-
Q. petraea	5856	8271±622	-	-	-	-
Q. pubecens	5959	-	-	-		3548±189
Q. ilex / Q. suber	18	-	-	144±20	-	-
Q. rubra	5761	-	4985±241	-	-	-

4.4 Definition of the bidirectional flux of isoprene degradation products

Our results show a negligible level of constitutive iox emission in unstressed plants, and in plants treated with high O_3 (Fig. 5). The uptake of iox increased linearly with exposure to increasing concentrations of ambient iox (from 0 to 6 ppbv of a 1:1 = MVK/MAC mixture) in all the investigated species, indicating iox fast removal and low compensation point in unstressed and stressed conditions. We conclude that plant capacity to take up iox should be included in global models that integrate estimates of iox formation, emission, and photochemical reactions in the atmosphere.

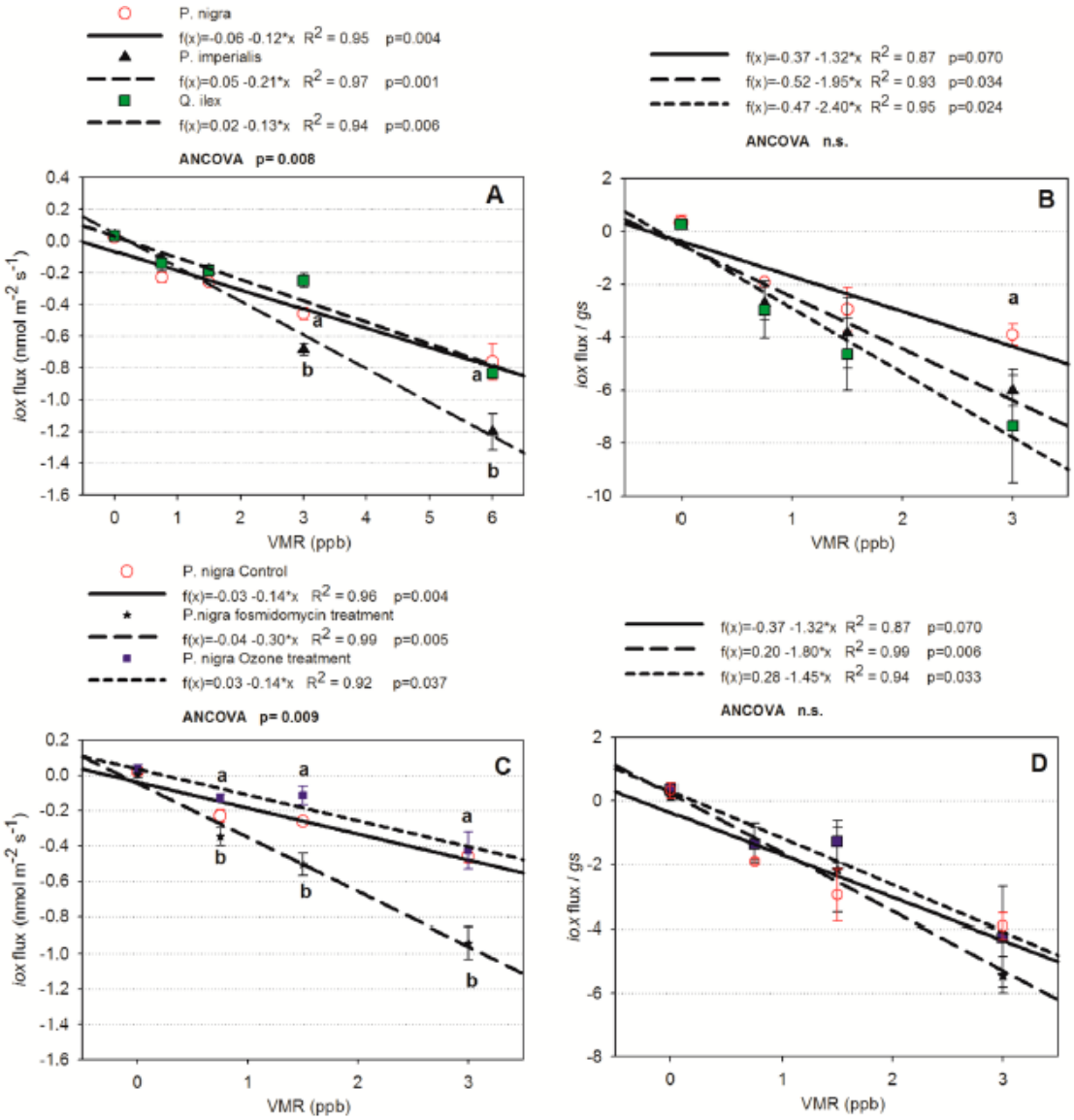

Fig. 5: (A) Flux of MVK+MAC (iox) at increasing iox concentration fumigated in the leaf enclosure for Paulownia imperialis, Quercus ilex, and Populus nigra under control conditions; (B) the same relationships shown in (A) but using data normalized for corresponding values of stomatal conductance (gs). (C) iox flux for control, fosmidomycin, and ozone-treated poplar leaves. (D) Normalization for corresponding values of gs. Different letters show significant differences at the same iox concentration (average ± SE, n = 7, Holm−Sidak method, $p < 0.05$). Levels of significance, p, are shown for linear regressions and analysis of covariance (ANCOVA), that were used for testing whether iox fluxes were correlated to air concentrations and whether the regression slopes are different from each other, respectively.

5. Publications:

PhD thesis, Cheng Wu: Emissions of biogenic volatile organic compounds and ozone balance under future climate conditions, 2015.

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Guenther, A. B., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.

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7. List of Documents/Annexes: