





INVESTICE DO ROZVOJE VZDĚLÁVÁNÍ

BVOC emissions and tropospheric ozone removal from crop and forest species

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Tato akce se koná v rámci projektu:

Vybudování vědeckého týmu environmentální metabolomiky a ekofyziologie a jeho zapojení do mezinárodních sítí (ENVIMET; r.č. **CZ.1.07/2.3.00/20.0246**) realizovaného v rámci Operačního programu Vzdělávání pro konkurenceschopnost.

Outline:

PART 1

Why study troposheric ozone?

What are Biogenic Volatile Organic Compounds (BVOC)?

PART 2

Experimental measurements: from leaves to ecosystem

Time scale sec hours days years We cover two of these important levels of investigation over three of the different time scales



leaf Canopy Landscape Regional/global Spatial scale

Ozone in low troposphere: an increasing threat for plants

Ozone in the stratosphere is GOOD!

protects life on Earth from the sun's harmful ultraviolet (UV) rays

Ozone in the troposphere is **BAD**!

Damages humans and plants, greenhouse gas



and factories all emit air pollution that forms ground-level ozone, a primary component of smog.

EPA (2010)



Relevance of plants for CO2 and atmospheric pollutant removal



1. Stomatal sink. Stomatal opening regulate carbon uptake and largely contribute to pollutant removal in the atmosphere

NO_x SO₂ O₃

CO2 uptake, photosynthesis. Detoxification of pollutants





2. Surface deposition on cuticles and soil. Adsorption processes

3. Chemistry in the gas phase. Reactions between BVOC and ozone

"Nonstomatal uptake"

Regulations to assess ozone risk to plants:

The present regulation in US and EU to assess critical ozone levels, is mostly based on estimates of an accumulated exposure over a threshold concentration (eg AOT40, SUM0)

Scientific consensus is that flux estimates are more accurate because they include analysis of plant physiology and different environmental parameters that control the uptake of ozone (not just the exposure)



UNECE Convention on Long-range Transboundary Air Pollution

MANUAL

on methodologies and criteria for

MODELLING AND MAPPING CRITICAL LOADS & LEVELS

and Air Pollution Effects, Risks and Trends

Ozone formation in the troposphere



What are plant volatiles ?

Volatile organic compounds (VOC)



"Living plants" Litter Vegetation burning Phytoplancton Soil microorganisms

Anthropogenic (AVOC)

Small scale combustion
Mobile sources
Solvent use
Fossil fuel production and distribution
Chemical industry
Fossil fuel combustion

Types of Biogenic VOC

- Hemiterpenoids $\rightarrow C_5$, only a few produced naturally
 - Isoprene



(C₅H₈, alkene)

Methylbutenol
 (C_sH₁₀O, alcohol)



- Monoterpenoids $\rightarrow C_{10}$, thousands of different structures
 - Limonene
 - myrcene
 (alkene)



- α- pinene (alkene)
- linalool (alcohols)



- Sesquiterpenoids $\rightarrow C_{15}$, most varied class of terpenoids
 - ß-caryophyllene

farnesene

Amount Known

- Isoprene (C₅H₈)
- Monoterpenes (C₁₀H₁₆)
- Oxygenated VOC
- Sesquiterpenes (C₁₅H₂₄)

The emission of BVOC from plants



Plants recycle some Carbon fixed with Photosynthesis to form BVOC!

The emission of BVOC from plants

Cytosol and Choloroplasts are the main site of production of isoprenoids



Isoprene (choroplastic origin) is more depending on photosynthetic pathway, while monoterpenes depend more on temperature-driven catabolism of sugars in the cytosol

Source of volatiles in the plant



Why BVOC are emitted?

Protection of the plant against:



Nature Chemical Biology

Plants interact with the ecosystem using BVOC





 Key component in the biosphere-atmosphere interactions.
 Sunlight

- Affect other plants and organism
- Favour the ecosystem perturbation

 Play an additional or alternative role in plant defences

BVOC & Ecosystem changes

The ozone-forming potential from certain VOC species has to be considered



Ozone is photochemically produced when VOC and NOx are abundant and around a ratio of 8!



$OFP_{species} = B[(E_{iso}R_{iso}) + (E_{mono}R_{mono})]$

where B is the biomass factor [(g leaf dry weight m⁻² ground area)], E_{iso} and E_{mono} are speciesspecific mass emission rates [(μg VOC) g⁻¹ leaf dry weight day⁻¹] for isoprene and monoterpenes respectively, R_{iso} and R_{mono} are reactivity factors [g O₃ g⁻¹ VOC].

Dust and aerosol primarily emitted from combustion sources and formed by photochemical activity

Rome, Italy

San Francisco, California, USA

Mexico City, Mexico



Beijing, China



VOC emission in changing ecosystems



Climate change & Species composition

Climatic areas for holm oak (Quercus ilex)

In southern and central Europe the most sensitive forest types are tree species which are at the southernmost limit of their distribution range. Thermophilous trees particularly Mediterranean **evergreen oaks** favored at the expense of conifers & beech



Environmental and Experimental Botany

journal homepage: www.elsevier.com/locate/envexpbot

The challenge of Mediterranean sclerophyllous vegetation under climate change: From acclimation to adaptation

Filippo Bussotti, Francesco Ferrini, Martina Pollastrini, Alessio Fini*

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Projection in 2100 is based on the ARPEGE model (Meteo-France) using a scenario with an increased mean temperature of 2.5 C.

(Badeau et al., 2007)

Climate change affects plant composition and VOC emission

Contraction of *P. sylvestris* at the lower limit of its range in Switzerland is in favor of a greater expansion of the deciduous xerotolerant oak species *Quercus pubescens*

-Global Change Biology

Global Change Biology (2013) 19, 229-240, doi: 10.1111/gcb.12038

Driving factors of a vegetation shift from Scots pine to pubescent oak in dry Alpine forests

ANDREAS RIGLING*, CHRISTOF BIGLER†, BRITTA EILMANN*¶, ELISABETH FELDMEYER-CHRISTE*, URS GIMMI*, CHRISTIAN GINZLER*, ULRICH GRAF*, PHILIPP MAYER‡, GIORGIO VACCHIANO§, PASCALE WEBER*, THOMAS WOHLGEMUTH*, ROMAN ZWEIFEL* and MATTHIAS DOBBERTIN*

Comparing the **altitudinal distribution** of 171 forest plant species between 1905 and 1985 and 1986 and 2005 along the entire elevation range (0 to 2600 meters above sea level) in west Europe



Species composition change in favour of high VOC emitter?

The greater terpene emission capacity may confer protection against multiple stresses and may partly account for the success of the **invasive species**, and may make invasive species more competitive in response to new global change-driven combined stresses

Global Ecology and Biogeography, (Global Ecol. Biogeogr.) (2010) 19, 863-874



Measurement of volatile terpene emissions in 70 dominant vascular plant species in Hawaii: aliens emit more than natives

Joan Llusià¹*, Josep Peñuelas¹, Jordi Sardans¹, Susan M. Owen² and Ülo Niinemets³

Ecosystem management & VOC emission

Abandonment of forest management favours senescent trees with low natural regeneration and loss of biodiversity







Contents lists available at ScienceDirect Agricultural and Forest Meteorology

ural and Forest Meteorology 192–193 (2014) 18–2



journal homepage: www.elsevier.com/locate/agrformet

Competition and tree age modulated last century pine growth responses to high frequency of dry years in a water limited forest ecosystem

Jaime Madrigal-González*, Miguel A. Zavala



Disturbances, ecological successions and structural changes



Successional dynamics affect species composition and structural changes of the canopy

Changing ecosystems in urban areas

Urban management plays an important role in determining future urban BVOC emissions in response to environmental changes



Role of Management Strategies and Environmental Factors in Determining the Emissions of Biogenic Volatile Organic Compounds from Urban Greenspaces

Yuan Ren, † Ying Ge, † Baojing Gu, ‡ Yong Min, $^{\$}$ Akira Tani, $^{\parallel}$ and Jie Chang*, †

A high ecosystem service value (e.g., lowest BVOC/leaf mass ratio) could be achieved through positive coping in confronting environmental changes and adopting proactive urban management strategies on a local scale, and optimizing the species composition of existing and newly planted trees



BVOC (RH)

Stressed ecosystems & emission changes

VOC emissions change in response to oxidative stress: the ecosystem may turn from source to sink

Plant, Cell & Environment

Plant, Cell and Environment (2014)

doi: 10.1111/pce.12322

Invited Review

Bidirectional exchange of biogenic volatiles with vegetation: emission sources, reactions, breakdown and deposition

Ülo Niinemets12, Silvano Fares2, Peter Harley1 & Kolby J. Jardine4





Can emission models properly take into account ecosystem changes? Are current vegetation inventories adequate?

Emissions based on three different vegetation inventories diverge due to a potential missattribution of broad-leaved trees and reduced tree-cover



Oberbolz et al. ACP 2013

(a) S1







Divergent Emission factors produce emission uncertainties

The uncertainties in emission estimates lead to uncertainties (2 to 10%) in life-time prediction of ozone, CO, CH4 Atmos. Chem. Phys., 13, 2857-2891, 2013 www.atmos-chem-phys.net/13/2857/2013/ doi:10.5194/acp-13-2857-2013 © Author(s) 2013. CC Attribution 3.0 License. Atmospheric Chemistry and Physics



Quantifying the uncertainty in simulating global tropospheric composition due to the variability in global emission estimates of Biogenic Volatile Organic Compounds

J. E. Williams¹, P. F. J. van Velthoven¹, and C. A. M. Brenninkmeijer²



Silvano Fares,^{†,*} Ralf Schnitzhofer,[‡] Xiaoyan Jiang,[§] Alex Guenther,[§] Armin Hansel,^{‡,||} and Francesco Loreto[⊥]

A global model (MEGAN coupled with CLM) run with realistic BEF from prevailing MT emitting species predicted lower concentrations of tropospheric ozone compared with an isoprene emitting scenario Integration of vegetation maps & national forest inventories: Emission Factor attribution for a realistic estimate

Do we have enough information on BEF? Is it still worth measuring BEF at leaf/ecosystem scale?





Climate change & biotic stress: overlapping signalling & VOC emission

Copolovici et al. 2014 J. Env. Exp. Bot.: Larvae feeding induced emissions of stress marker compounds (*E*)-β-ocimene and homoterpene DMNT and methyl salicylate from *Alnus glutinosa*. The emissions were more strongly elicited in drought-stressed plants.



Winter et al. PCE 2012: overlapping signalling and responses to abiotic and biotic stress factor demonstrated by stronger expression of herbivore-induced volatiles by high levels of heavy metals

Part 2: Laboratory studies help understanding the net balance between ozone formation by BVOC and sequestration, and the antioxidant role of BVOC

Methods: enclosures

In a greenhouse or in a lab, where is possible to control environmental parameters, leaf/branch enclosures allow measuring gas exchage according to the Fick`s law:



F=air Flux A^L =Leaf AreaX = Concentration of CO2, Water, ozone, BVOC



Flux intercomparisons



Results: ozone uptake from plants



10

0

201/106 27.3¢

60.00 SOLLES

13.1.0° 2.12

PAR

13,106 9.36

13-1106 12:00

P3-11/06 TAR 20/1/24

133,7106,76,80

Loreto and Fares, Plant Physiology 2007

Isoprenoids = antioxydants

100 = % ozone damage normalized vs plants which do no emit isoprenoids



Leaf which

naturally

produces

isoprene

Isoprene production was inhibited

Where does the reaction between ozone and BVOC take place?

C-Labeling experiments suggest that isoprene reacts with ROS in leaves, and emission of its reaction products is directly proportional to heat stress



Global Change Biology (2012) 18, 973-984, doi: 10.1111/j.1365-2486.2011.02610.x

Within-plant isoprene oxidation confirmed by direct emissions of oxidation products methyl vinyl ketone and methacrolein

KOLBY J. JARDINE*, RUSSELL K. MONSON†, LEIF ABRELL‡§, SCOTT R. SALESKA¶, ALMUT ARNETH**††, ANGELA JARDINE*, FRANÇOISE YOKO ISHIDA‡‡, ANA MARIA YANEZ SERRANO‡‡, PAULO ARTAXO§§, THOMAS KARL¶¶, SILVANO FARES***, ALLEN GOLDSTEIN†††, FRANCESCO LORETO‡‡‡ and TRAVIS HUXMAN*¶



Which role of BVOC?

Holm oak is a strong monoterpene emitter = = major role of BVOC in removing ozone!



stomatal conductance to ozone

Laboratory experiments at Forschungszentrum-Juelich: testing O3 formation of α-pinene under NOx limited conditions

~75 ppb NO_x


Part 2: Field measurements to test the capacity of VOC-emitting trees to remove pollutants

A canopy-scale approach: Eddy Covariance flux measurements



CO2 flux: Subtracting modelled ecosistem respiration to the Net Ecosystem Exchange (NEE) , Gross Primary Productivity (GPP) is calculated

Water flux: Stomatal conductance is calculated from measured transpiration by inversion of Monteith equation, therefore an estimate of stomatal ozone fluxes is possible

Ozone fluxes: sum of stomatal and non stomatal components

How is ozone deposition partitioned between various sinks?

Rvoc???

BIG LEAF MODEL:

A series of resistances (aerodynamic, boundary layer, stomata) reduce flux magnitude from the atmosphere to inside the leaves, obeying Ohm's law.

$$F_{O3} = F_{O3sto} + F_{O3nsto} = \frac{[O_3]_c}{R_{sto}} + \frac{[O_3]_c}{R_{nsto}}$$

Monteith equation to calculate Stomatal Resistance, when latent heat is measured



Partitioning ozone fluxes among its various sinks: stomata

Eddy Covariance above canopy



Partitioning ozone fluxes among its various sinks: stomata

Monteith equation to calculate stomatal conductance

After subtracting the evaporative component



Most of ozone removed by forest is in the canopy region, a portion up to 70% is removed by stomata!

Eddy covariance data to parameterize Ball-Berry equation

FOR A SINGLE LEAF

 $G_{sto} = \frac{m^* A^* R H}{Ca} + G_0$

FOR A CANOPY

 $f(H) = \frac{Ca * F_{LE}}{v * \delta * L * \rho_a * (q_{sat(T_a)} - q_a) * A}$

The relationship between RH and Gsto is not linear, modification of m (=f(H)) calculation produces the best agreement with observed Gsto

 $Ca * G_0$

 $a^*b\exp(RH)^*$

 $+G_{0}$



Modelled stomatal conductance: the DO3SE approach

$$G_{sto_{DOSE}} = g_{\max} * f_{phenol} * f_{light} * \max\{f_{\min}, (f_{temp} * f_{VPD} * f_{SWP})\}$$

Calculation of non-stomatal ozone fluxes by BVOC

Calculation steps: **1) Characterization of the reacting BVOC species :**

Fluxes of VOC species can be measured with GC-MS exposing branches in enclosures to ozone-free air under standard environmental conditions, thus obtaining Basal Emission Factors

2) Fluxes are then modelled using the algorithms proposed by Tingey et al. (1993) and Guenther et al. (1995) in which BVOC emission are function of light and temperature:

$$E_{L+T} = BEF\left[\frac{\alpha C_L PAR}{\sqrt{1+\alpha^2 PAR^2}}\right] * \left[\frac{\exp\left(\frac{C_{T1}(T-T_s)}{RT_s T}\right)}{0.961 + \exp\left(\frac{C_{T2}(T-T_M)}{RT_s T}\right)}\right]$$

3) Reactivity with ozone: we can assumed 1 mol (O₃) reacting with 1 mol (BVOC), considering past experiments using smog chambers

$$E_T = BEF \exp\left[\beta \left(T - T_s\right)\right]$$





Non-stomatal ozone fluxes: NO

Definition of a BEF for NO according to Firestone et al. (1986) and Steinkamp et al. (2010):

$$F_{NO(10^{\circ} < T \le 10^{\circ}C)} = 0.28 \cdot T_{soil} \cdot BEF; \qquad F_{NO(10^{\circ} < T \le 30^{\circ}C)} = e^{0.103T_{soil}} \cdot BEF; \qquad F_{NO(T > 30^{\circ}C)} = 21.97 \cdot T_{soil} \cdot BEF$$

NO flux from soil was modeled for all year and we assumed 1 mol (NO) reacting with 0.8 mol (O_3), accounting for the *uv*-driven O_3 formation due to the conversion of some NO molecules in NO₂ (Dillon et al. 2002).



Other non-stomatal ozone fluxes

Cuticular resistance as function of humidity and turbulence (Zhang et al. 2002; Erisman et al. 1994)

 $Rcut(dry) = \frac{Rcut(dry)_0}{e^{0.03RH} \cdot LAI^{1/4} \cdot u_*} \qquad Rcut(wet) = \frac{Rcut(wet)_0}{LAI^{1/2} \cdot u_*}$

fc

Below-canopy resistances as the sum of in-canopy aerodynamic resistance and ground resistance as function of LAI, tree height, turbulence and soil water content, all parameters measured on site (Zhang et al. 2002, Bassin et al. 2004)

$$Rac = \frac{14 \cdot LAI \cdot zc}{u_*} \qquad \qquad Rg = 200 \cdot 300 \cdot \frac{SW}{SWC}$$



Evidences from field experiments: the Holm oak urban forest in Castelporziano

Above the canopies of Mediterranean oaks and pines a complex photochemistry takes place with concurrent phenomena of ozone formation and ozone deposition





Biogeosciences, 6, 1043–1058, 2009 www.biogeosciences.net/6/1043/2009/ © Author(s) 2009. This work is distributed under the Creative Commons Attribution 3.0 License. ~ 6000 ha, 25 km from Rome downtown

The ACCENT-VOCBAS field campaign on biosphere-atmosphere interactions in a Mediterranean ecosystem of Castelporziano (Rome): site characteristics, climatic and meteorological conditions, and eco-physiology of vegetation

S. Fares^{1,2}, S. Mereu³, G. Scarascia Mugnozza¹, M. Vitale³, F. Manes³, M. Frattoni⁴, P. Ciccioli⁴, G. Gerosa⁵, and F. Loreto¹

Atmospheric Environment 67 (2013) 242-251

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment



Biogeosciences

journal homepage: www.elsevier.com/locate/atmosenv

Testing of models of stomatal ozone fluxes with field measurements in a mixed Mediterranean forest

S. Fares ^{a.} ^{*}, G. Matteucci ^{b.c}, G. Scarascia Mugnozza ^a, A. Morani ^b, C. Calfapietra ^{b, f}, E. Salvatori ^d, L. Fusaro ^d, F. Manes ^d, F. Loreto ^e

Ozone fluxes in the Holm Oak forest

Ozone fluxes are higher during warm days, when non-stomatal sinks (e.g. gasphase chemistry) are higher. Up to 8 g O3 m-2 are sequestrated every year!



Atmospheric O3 concentration gradient from the oil to above the canopy





Fares et al. Atm. Env. 2013; Fares et al. Agr. For. Met. 2014; Savi et al. 2014

Clear diurnal fluxes of isoprene, monoterpenes and of Methyil Vinyl Chetone (MVK), one of the main oxidation products of isoprene



Davison et al. Biogeos. 2009; Fares et al. Biogeos. 2009

Evidences of non-stomatal ozone fluxes: VOC concentrations

Norm. concentrations and meteor. variables



Sesquiterpenes react with ozone in few seconds, therefore they disappear fast during the day when atmospheric ozone builds up

Non-stomatal ozone fluxes peak in coincidence with the highest concentration of Methyl-vynilketone + Metacrolein, two oxidation products from reaction between ozone and isoprene

SQT>>Isoprene>>Acetone

The field campaign in 2014

Preliminary data show how Holm Oak is a predominant Monoterpene emitter Deployment of PTRMS to the field site









Fluxes peak during the day because primary emitted BVOC depend on light and temperature

Not only O3 & BVOC: urban Oaks of Castelporziano can remove PM and NOx from the atmosphere!





field measurements in Exeter(Central valley, CA)















Views of Research Site

Seatainer housing instruments



Tower







Stomatal ozone flux is a minor fraction of total ozone flux

. 4

Ozone fluxes - daily average

Fares et al. Env.

Poll. 2012



BVOC concentration in the Orchard:

Fares et al. ACP 2012



Isoprenoids are emitted during all year



Monoterpenes and isoprene are emitted from the plants as a function of light and temperature, but still air in the evenings causes them to accumulate to higher concentrations (highest near the ground). Fares et al. ACP 2012 During all year, the orchard is a source of benzenoid compounds



Benzenoids are in floral scents, but are emitted all the year even if we don't smell them!

Fares et al. ACP 2012

BVOC fluxes



Large fluxes during the flowering period...

Fares et al. ACP 2012

Stomatal and non-stomatal fluxes: final balance



We believe that the model **underestimates Fvoc** during **flowering**, because not fully accounting for the burst of BVOCs coming from flowers.

Fares et al. Env. Poll. 2012

Long term field measurements in Blodget Forest

Since 1999, Eddy covariance and gradient measurements of ozone, CO2, have been performed

Through a multiyear analysis (2001-2006), we observed different sinks of ozone uptake, elucidating their dependence on plant physiology and environmental conditions





Daily ozone concentration & fluxes

The daily ozone concentrations showed maximum diurnal peaks above 90 ppb!



Stomata represented a significant sink of ozone for this ecosystem, but not the major sink, similarly to Citrus site!



Fares et al. Agr. For. Met. 2010

Evidence of non-stomatal ozone removal at Blodgett:



the day in coincidence with monoterpene fluxes and the fluxes of the oxidation products of monoterpenes (m113)!

We calculated that up to 40 % of ozone uptake is due to BVOC!

Non-stomatal ozone fluxes

How much ozone is removed every year?



About 7 grams of ozone are removed per square meter of orchard, comparable to a ponderosa pine plantation located in the Sierra Nevada Mountains.

The Citrus orchard and Pine plantation have approximately the same efficiency for ozone removal, since cumulated ozone concentration is similar at these sites.



Conclusion form field studies (II): Ozone impact on GPP

- Reduction in carbon assimilation was more related to stomatal ozone flux than to ozone concentration.
- The negative effects of ozone occurred within a day of exposure/uptake



Fig. 3 Wavelet coherence analysis to look the temporal correlations between the residuals of gross primary productivity (GPP) and ozone concentration (a, c) or stomatal ozone deposition (b, d) for the Castelporziano site. The colors for power values are from blue (low temporal correlations with GPP) to red (high temporal correlations with GPP). The thick black line in a and b indicates the cone of influence that delimits the region not influenced by edge effects. Days of the year (DOY), days after September 1st of year 2011.

Fares et al (2013) GCB

Conclusion form field studies (II): Ozone impact on GPP

Long-term continuous eddy-covariance measurements (>9 years at 30 min resolution) at three Mediterranean-type sites showed that Up to 12–19% of the carbon assimilation reduction is explained by higher stomatal ozone flux!

Random Forest Analysis of the effects on GPP at three Mediterranean-type ecosystems: Pinus ponderosa, Citrus sinensis, Quercus ilex





Semi-empirical models for the assessment of ecosystem services provided by urban trees

Remote sensing and modeling



Measurements on site



Literature





Bibliography: Goudriaan and van Laar, 1994; Stavrakou et al., 2008; Farquhar et al., 1980; Baldocchi, 1994; Harley et al., 1992; Ball et al., 1987.

Model validation (in progress)







Field measurements are used to validate modelled data







Conclusions

- Comparing VOC emissions and O₃ uptake is important for studying the role of plants for tropospheric O₃ balance.
- Future scenarios of climate change may affect species composition and BVOC emission.
- The O₃ forming potential of VOCs highly depends on NO_x concentration. As long as air is polluted, strong emitters tend to be a source of ozone. It is worth paying attention to consider the species with high emission rates as sources of ozone in urban or suburban area.
- Modelling is the way to assimilate experimental results and provide urban planners a valuable tool to select the most suitable tree species which maximize ecosystem services. However, extensive parameterization is still needed!






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Thank You!

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Tato akce se koná v rámci projektu:

Vybudování vědeckého týmu environmentální metabolomiky a ekofyziologie a jeho zapojení do mezinárodních sítí (ENVIMET; r.č. **CZ.1.07/2.3.00/20.0246**) realizovaného v rámci Operačního programu Vzdělávání pro konkurenceschopnost.