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Supplementary Materials for

Ozone affects plant, insect, and soil microbial communities: A threat to terrestrial ecosystems and biodiversity

Evgenios Agathokleous*, Zhaozhong Feng*, Elina Oksanen, Pierre Sicard, Qi Wang, Costas J. Saitanis, Valda Araminiene, James D. Blande, Felicity Hayes, Vicent Calatayud, Marisa Domingos, Stavros D. Veresoglou, Josep Peñuelas, David A. Wardle, Alessandra De Marco, Zhengzhen Li, Harry Harmens, Xiangyang Yuan, Marcello Vitale, Elena Paoletti

*Corresponding author. Email: evgenios@nuist.edu.cn (E.A.); zhaozhong.feng@nuist.edu.cn (Z.F.)

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Ozone in the atmosphere

Higher solar radiation, biogenic VOC emission, transport of O_3 from stratosphere to troposphere, and lower O_3 removal by deposition and reactions with NO explain the highest O_3 burdens observed at high elevation (*206*, *207*). Over Greenland and Northeast Siberia, the high O_3 levels can be attributed to high insolation and high levels of reactive compounds (e.g. oxidized nitrogen species) present in the surface layer during the sunlit period due to local sources (e.g. NOx emissions from snowpack, PAN decomposition, boreal forest fires) and large-scale O_3 transport and stratospheric O_3 inputs (*9*, *208–210*). Around the Mediterranean basin, high AOT40 values are mainly due to industrial development, road traffic increment, high insolation, sea-land breeze recirculation and long-range transport of O_3 precursors and O_3 (*199*).

Ozone-induced visible foliar injury

It is well documented that O_3 can cause severe visible injury to exposed plants (*211–222*). The severity and the distribution of injury to leaves and plants, as well as the appearance (stippling, flecking, bronzing, mottling, chlorosis, discoloration and necrosis) of injury depend on plant species, physiological state, environmental conditions, and duration and nature (acute or chronic) of exposure (*214*). Acute exposures are characterized by high O_3 concentrations for a relatively short time, restricted to hours or days, while chronic exposures involve lower concentrations that persist or recur over a period of weeks or months (*221* *223*). Stippling, flecking and bifacial necrosis are common visible foliar injuries produced by acute O_3 exposure (*221*, *224–226*), while chlorosis (yellowing or paling of green leaves), upper-surface stippling, premature senescence and necrosis (*224*, *227*, *228*) are commonly produced by chronic O_3 exposure.

In broadleaved species, visible foliar O_3 injury is limited to the upper leaf surface, generally categorized as stipple, chlorosis, or fleck (*229*). Bifacial necrosis may occur at later stages of development. Injury is more severe on mid-aged and older leaves than on younger leaves (*224*). Sometimes visible injury occurs as a general discoloration, reddening or bronzing between the leaf veins. Stippling is characterized by interveinal, dot-like areas of tan, red, brown, purple or black pigmentation on the upper surface of the leaf (*230*). The production of stipple requires exposure of the leaf surface to direct sunlight. An overlapping leaf can protect the lower leaf producing a "shadow" in the injury on the leaf surface. Chlorosis is a loss of chlorophyll in the leaf (non-green pigmentation) and appears in relatively discrete patches on the leaves known as mottles. Fleck is characterized by small, discrete areas of dead tissue in the palisade mesophyll. Lesions may be irregular in shape and range in color from tan to black. Bifacial necrosis is a more severe form of injury resulting from cell death in both the palisade and spongy mesophyll and epidermal tissues. Injury appears on both sides of the leaf and dead tissue may take on a papery texture. Color can range from light tan to black.

Ozone injury on conifer needles generally appears as tipburn or chlorotic mottling. Tipburn is considered to be primarily induced by acute exposure and mottling by chronic exposure. Injury usually progresses from the needle tip to the base (*231*, *232*). Tipburn may become red to brown. Chlorotic mottling is the most common visible injury for conifers. Mottle can be described as discrete patches of similar size, yellow or light green, without sharp borders with the green zones. Chlorotic mottling seems to increase with increasing needle age (*216*, *233*, *234*). Furthermore, mottling is more distinct on light-exposed needle areas than on shaded ones.

Visible foliar O_3 injury can be differentiated from symptoms induced by other biotic and abiotic stressors occurring at the same area (*219*, *235*, *236*). Ozone-induced injury can be differentiated from fungi and insect damage by in-hand observation of color and pattern of the symptoms (*224*, *228*, *235*, *237*). Careful observation of chlorotic tissue often reveals a distinct necrotic point at the center of the discolored area, where the insect penetrated the epidermis with its piercing mouth parts (*231*). Winter fleck, caused by snow and cold temperatures, is a symptom that superficially resembles mottling. These lesions are roundish with distinct margins, tan or brown, and vastly more abundant on the adaxial surface of the needle, suggesting the role of sunlight in their formation (*231*). The winter flecks on older needles were elucidated at the cell level (*236*). An injury initially caused by O_3 then aggravated during winter, presumably following the formation of ice crystals at the most damaged locations. The injury was likely further intensified during the following growing season as a consequence of steady oxidative stress at the periphery of the symptomatic area. Needle injury from road salt accumulation, drought, desiccation, winter injury and lightning can be visually distinguished from O_3 injury by the color, shape, pattern of development on the foliage, and occurrence in the tree crown (*232*, *238*).

Intensive research over the last 10 years has led to significant databases about the effects of O_3 exposure on the cell structure both in conifers (particularly *P. cembra* and *P. halepensis*) and broadleaf species (*211*, *213*, *215*, *219*, *232*, *239*, *240*). Microscopic investigation can be used to validate visible O_3 injury (*240*, *241*).

A first microscopic exam allowed the characterization of the injury: localization, extent, color, and presence of biotic marks. After the correct identification of the suspected injury, a validation was carried out by microscopy on doubtful samples collected during the field survey. The typical ozone injury occurred only in the palisade mesophyll, with lysis followed by the collapse of cells, with tannins and anthocyanin accumulation producing a red-brown necrosis (*215*, *219*, *240*, *242*, *243*).

Ozone effects on plant communities

Table S1 summarizes studies on the responses of communities to O_3.

Global maps of AOT40 and endemism richness

The areas of high endemism richness of vascular plants (> 200 species per 10,000 km²) are: Northern Hemisphere Atlantic islands at high O_3 risk in the latitude band 15-45°N (e.g. Canary islands, Azores and Caribbean), Pacific Islands (Polynesia-Micronesia, Melanesia, Philippines) and Indian Islands (e.g. Madagascar, Seychelles). For the latitude band of the Atlantic islands, the reader may refer to (*9*, *244*, *245*); the tropospheric warming is stronger in the latitude band 15-45°N (*246*). Islands are well-known centers of high levels of endemism, represent 3.6% of the terrestrial surface, and have 26.1% of all worldwide vascular plants (Kier et al., 2009). Among the Islands, Tonga, Indonesia, Japan, New Zealand and Tasmania have lower endemism richness (60-80 species per 10,000 km²). From a global perspective, the island of New Caledonia had by far the highest value with 1,350 vascular plant species per 10,000 km^2 (*197*). Regarding mainland regions, the endemism richness hot-spots (>50 species per 10,000 km^2) are observed in California, Tropical Florida, Central and Western South America (Chile, Ecuador and Peru), Guyana, Brazilian coastline, Ethiopia, South Africa, Himalayan and mountains of Southwestern China, Southeastern and extreme Southwestern Australia and around the Mediterranean basin (Fig. 3). If the threshold is set to 20 species per 10,000 km^2, South America, Equatorial Africa and South Asia are considered hot-spots too. Among mainland areas, regions with Mediterranean-type climate emerge as global centers of plant endemism richness (*197*). The South African Cape region hosts high endemism richness (771 species per 10,000 km^2), ranking second among all 90 biogeographic regions.

Table S1. Studies on the responses of communities to elevated ozone (O_3). “g.s.” stands for growing season. AOT40 is the sum of the positive differences between the hourly mean O_3 mixing ratio and the threshold of 40 ppb multiplied by the 1-h averaging time, during daylight hours in a defined growing season, depending on the type of vegetation (*247*).

Study No	**Community**	**Origin of material**	**Community description**	**Duration of exposure**	**Ozone exposure**	**Main results**	**Reference**
Open-top chambers (OTCs)							
1	Turfs of calcareous grassland.	Twyford Down, Hampshire (51^{o}02’42’’N, 1^{o}18’33’’W).	Intermediate between the National Vegetation Classification communities CG3d *Bromopsis erectus* grassland *Festuca rubrae-Festuca arundinacea* sub-community (EUNIS2004 E1.26 Sub-Atlantic semi-dry calcareous grassland) and MG1 *Arrhenatherum elatius* grassland *F. rubra* sub-community (EUNIS2004 E2.22 Sub-Atlantic lowland hay meadows).	3 g.s.	Mean seasonal AOT40≈7.4-18.8 ppm h.	One species was lost from the O_3 treatments, but not the control, and the overall effects on species composition were relatively small.	(*51*)
2	Artificial communities of species typical of calcareous grasslands	A chalk grassland site in SE England.	‘Each mixture contained the same forb species (*Campanula rotundifolia*, *Leontodon hispidus*, *Lotus corniculatus* and *Sanguisorba minor*), but one half had three grass species typical of swards dominated by *Festuca ovina* (*Briza media*, *Koeleria macrantha* and *Festuca ovina*), while the other half of the pots had three grass species typical of swards dominated by	3.5 months	Filtered air, or filtered air with added 50, 70 or 90 ppb O_3 during the	Elevated O_3 changed species composition of the artificial communities, with less forbs with increasing O_3 concentrations above 40 ppb. Elevated O_3 also resulted to more calcareous grassland	(*47*)

	as well as semi-natural chalk grassland swards.		*Festuca rubra* (*Bromopsis erectus*, *Dactylis glomerata* and *Festuca rubra*).'		daytime.	communities in the semi-natural swards.	
3	Managed pasture composed of grasses, weeds and clover.	A commercial seed company.	Standard seed mixture no. 330 (species not detailed).	2 g.s.	O_3 concentrations 0.50, 0.85, 1.11 and 1.64 times the O_3 concentrations in the ambient air.	Negligible shifts in species composition, a result that might have occurred due to experimental artifacts.	(*55*)
4	Productive meadow community with two species.	n/a	A mixture of ladino clover (*Trifolium repens* cv. Regal) and tall fescue (*Festuca arundinacea* cv. Kentucky 31).	2 g.s.	O_3 concentrations 1.3-1.9 times the ambient O_3 concentration.	Reduction of ladino cover fraction to tall fescue.	(*59*)
5	Early successional plant community.	A research site on the Auburn University (Alabama) campus.	Most prevalent species included: blackberry (*Rubus cuneifolius*), broomsedge bluestem (*Andropogon virginicus*), bahia grass (*Paspalum notatum*), and *Panicum* spp. Winged sumac (*Rhus copallina*).	2 g.s.	O_3 concentrations 0.5 (filtered), 1.0 and 2.0 times the ambient O_3 concentrations.	Changes in the community structure, with lower species richness, diversity and evenness in the ambient and elevated O_3 treatments than the filtered air; the species dominating canopy cover was different between the filtered air (winged sumac) and the largest O_3 exposure treatment (blackberry).	(*49*)
6	Semi-natural temperate grassland.	Silwood Park, near Ascot, 32 km west of London. 'The experimental site was an area of semi-natural grassland classified as an *Arrhenatheretum elatius-Festuca rubra* subcommunity (National Vegetation Classification).'	'The major component species were *Agrostis capillaris*, *Festuca rubra*, *Poa pratensis*, *Veronica chamaedrys*, *Trifolium repens*, *Plantago lanceolata*, and *Stellaria gramineae*.'	2 g.s.	Ambient air pollution exposure (attributed to O_3 fluctuations).	Forb and legume fraction increased.	(*248*)

7	Weed community exposed previously to filtered air, 90 ppb O_3 or 120 ppb O_3, with episodic patterns of varying daily peak concentrations, for four growing seasons.	Weed populations were selected in a long-term O_3-exposure experiment carried out in Corvallis, Oregon, at the US Environmental Protection Agency Laboratory, Western Ecology Division. The seed banks were transferred to Brazil for experiments.	Twenty species have been identified at different densities in each season: *Spergula arvensis*, *Calandrinia ciliate*, *Medicago lupulina*, *Vicia tetrasperma*, *Rumex crispus*, *Polygonum persicaria*, *Taraxacum officinale*, *Digitaria sanguinalis*, *Erodium cicutarium*, *Solanum sarachoides*, *Raphanus sativus*, *Echinochloa crusgalli*, *Chenopodium album*, *Trifolium repens*, *Polygonum argyrocoleon*, *Datura stramonium*, *Convolvulus arvensis*, *Tanacetum vulgare*, *Oxalis corniculata*, and *Sonchus oleraceus*.	2 g.s.	Low pollution in two separate experiments.	Although the species richness of the communities for different O_3 exposure history was similar, the relative abundance of the dominant species was different among the communities; the annual species *Spergula arvensis* dominated the communities from 90 ppb and *Calandrinia ciliate* dominated the communities from 120 ppb.	(*50*)
8	Frequently cut mixtures of ryegrass and white clover.	n/a	Seeds of white clover (*Trifolium repens* cv. Ladino California) and perennial ryegrass (*Lolium perenne* cv. Bastion) were sown at a weight ratio of 1 : 6.	4 g.s.	Different O_3 exposure regimes in each season, in the range of ≈0.6-1.5 times the O_3 concentration of ambient air.	Clover fraction (in terms of biomass) over ryegrass was reduced by elevated O_3 but only in the first growing season.	(*58*)
9	Mixture of six species representative of annual pastures.	n/a	Six representative annual species were sown at a density of 1000 seeds m^{-2}: 3 legumes (*Trifolium striatum*, *Trifolium cherleri*, *Ornithopus compressus*), 2 grasses (*Briza maxima*, *Cynosurus echinatus*), and 1 herb (*Silene gallica*).	39 days	Filtered air, ambient O_3, ambient O_3 + 20 ppb and ambient O_3 + 40 ppb.	Legumes were found sensitive to O_3. In terms of biomass, *Ornithopus* exhibited positive effects whereas *Trifolium* species negative effects, thus, *Ornithopus* took advantage of clover species. This study also showed that within-family competition (here legume) can play the major driver of the overall response of a community.	(*42*)
10	a) Natural grassland community of *Trisetum flavescens* ×	A commercial seed bank (Schweizer AG, Thun, Switzerland).	*Trisetum flavescens* was planted in mixture (mixing proportions: 100:0, 75:25, 50:50, 25:75, and 0:100) with *Centaurea jacea* and with *Trifolium pretense*.	1 g.s.	Seasonal episodic exposures (AOT40=8.0-35.5 ppm h).	a) Reduced fraction of clover, in terms of biomass per area, after seasonal episodic exposures at the highest exposure (AOT40=35.5 ppm h) and in moist conditions, but not at	(*62*)

	Trifolium pretense and b) *T. flavescens* grown in a community with *Centaurea jacea.*					the medium exposure (AOT40=14.4 ppm h) or in dry conditions. b) When *T. flavescens* was grown in a community with *Centaurea jacea*, the fractions were not affected significantly by O_3 (AOT40=8.0 or 28.0 ppm h).	
11	Northern European lowland hay meadow community.	'Commercial seeds (Kukkiva Niitty) collected from natural populations in south-western Finland.'	'Large mesocosms with species typical to lowland hay meadows (EU Council directive 92). The mesocosms were designed to hold the major functional types of plants that occur on northern European lowland hay meadows: grasses (*Agrostis capillaris*, *Anthoxanthum odoratum*), herbs (*Fragaria vesca*, *Campanula rotundifolia*, *Ranunculus acris*) and legumes (*Trifolium medium*, *Vicia cracca*).'	3 g.s.	Ambient or elevated O_3 (40-50 ppb; 1.5 × ambient O_3 concentration).	While the community exhibited reduced aboveground biomass and four out of seven species exhibited reduced growth and/or visible injuries in response to elevated O_3, the dominance of different functional groups was not altered by elevated O_3.	(*249*)
12	Grass-clover mixture pasture.	n/a	A mixture of 15% (in terms of seed weight) red clover (*Trifolium pratense* cv. 'Fanny'), 60% timothy (*Phleum pratense* cv. 'Alexander') and 25% fescue (*Festuca pratensis* cv. 'Sval~Sfs Sena').	2 g.s.	Filtered air, ambient O_3 (11.8 and 5.1 ppm h in the first and second growing seasons) or O_3 concentrations ≈1.5 and 2.0 times those in the ambient air.	No significant result suggesting changes in species composition; only a chamber effect was found compared to open plots	(*250*)
13	A mixture of *Trifolium repens* (var. Grasslands Huia) and *Lolium*	n/a	A mixture of *Lolium perenne* var. Melle (25 kg seed ha^{-1}) and *Trifolium repens* var. Grasslands Huia (5 kg seed ha^{-1}).	22 days (July-September)	O_3 episodes (4–7 h concentrations of 50–70 nl l^{-1}) and ambient air	Herbage composition was significantly affected by O_3. Most yield losses were because of less *Trifolium*, which was more susceptible than *Lolium*.	(*56*)

	perenne (var. Melle).				(7-hour mean of 10–28 nl 1^{-1}).		
Solardomes							
14	A grassland species mixture with *Leontodon hispidus* grown with *Anthoxanthum odoratum* or *Dactylis glomerata*.	Commercial plug plants (British Wildflower Plants, Norfolk, UK).	Four plants of *Leontodon hispidus* and three plants of the grass species (*Dactylis glomerata* and *Anthoxanthum odoratum*).	5 months	Eight background O_3 concentrations ranged 21.4-102.5 ppb	Cover increased with increasing O_3 exposure for both *A. odoratum* and *D. glomerata*, increasing the grass:*Leontodon* cover ratio in both community types.	(*33*)
15	Mesocosms representing the BAP Priority habitat 'Calcareous Grassland'.	Commercial plug plants (British Wildflower Plants, Norfolk, UK).	Each mesocosm had 2 plugs per the species *Briza media* and *Festuca ovina*, and 1 plug per the species *Campanula rotundifolia*, *Sanguisorba minor*, *Scabiosa columbaria*, *Helianthemum nummularium* and *Lotus corniculatus*.	2 g.s.	Eight O_3 profiles for twelve-weeks in each g.s. (24 h mean = 15.6-73.0 ppb in the first year and 19.0-73.3 ppb in the second year)	Reduction in flower numbers was found in *Campanula rotundifolia* and *Scabiosa columbaria*.	(*45*)
FACE							
16	Subalpine grassland.	A typical pasture community in the subalpine zone of the European Alps and Pyrenees (EUNIS classification 35.1,http://eunis.eea.europa.eu/)	'The most frequent species are *Festuca violacea*, *Nardus stricta* and *Carex sempervirens*, which together account for approximately one-half of the cover. The remaining half is composed of > 70 forb and a few legume species.'	3 g.s.	O_3 concentrations 1.6 times the ambient ones in three growing seasons (AOT40=48.0 ppm h).	No significant effect on the number of species per monolith and the Shannon diversity index.	(*63*)
17	Semi-natural grassland.	A site at Le Mouret (754 m a.s.l., 46^{o}45'N/7^{o}10'E), Switzerland.	'The plant species composition was identified as an *Arrhenatherion elatioris* alliance…, confirmed by the presence of *Bromus hordeaceus*, but without a single dominant species. Important subdominants	5 g.s.	O_3 concentrations approximately 1.5 times	Reduced productivity, in terms of annual dry matter yield, with accompanied shifts in the fractions of functional groups, i.e. grasses, legumes, forbs; legumes fraction	(*251*)

			included the grasses *Holcus lanatus*, *Trisetum flavescens*, *Alopecurus pratensis* and *Arrhenatherum elatius*, the nonleguminous forbs *Plantago lanceolata* with *Ranunculus friesianus* and the legume *Trifolium pratense*. A total of 53 species of vascular plants was identified.’		the ambient ones over five growing seasons (AOT40=34.0 ppm h).	appeared more susceptible.	
18	Upland mesotrophic grasslands.	A site located in northern England at High Keenley Fell, about 18 km from Hexham (national grid reference NY 7922 5586 altitude = 360 m).	A *Festuca rubra–Holcus lanantus–Anthoxanthum odoratum* grassland. Components included: *Cerastium arvense*, *Cerastium fontanum*, *Chrysosplenium oppositifolium*, *Conopodium majus*, *Lathyrus pratensis*, *Leontodon hispidus*, *Plantago laneolata*, *Polemonium caeruleum*, Ranunculaceae, *Rhinanthus minor*, *Rumex acetosa*, *Taraxacum* spp.,*Trifolium pratense*, *Trifolium repens*, *Stellaria graminea*, *Veronica chamaedrys*, *Vicia cracca*, *Vicia hirsuta*, *Vicia sativa*, *Vicia sepium*.	2 g.s.	Ambient O_3 elevated by mean concentrations of only approximately 4-10 ppb.	Elevated O_3 reduced total biomass of herbs but not of grasses or legumes and affected species composition within the combined herb and legume component but not within grasses component.	(*41*)
19	Intact subalpine grassland.	A site in the Central Grisons Alps at Alp Flix, Sur, Switzerland (2000 m a.s.l., 9°39’N/46°32’E).	‘A typical species-rich pasture community in the tree line zone of the European Alps and Pyrenees, characterized among others by *Nardus stricta*, *Carex sempervirens*, *Arnica montana*, and *Gentiana acaulis*.’	7 g.s.	Ambient O_3 or O_3 concentrations 1.2 and 1.6 times those of ambient O_3.	Elevated O_3 did not affect the functional group productivity, and, among 17 species tested, only increased the abundance of *Nardus stricta*, a slow-growing species with a low specific leaf area; even this resulted to competitive advantage, it was not at the expense of other species abundance.	(*52*)
Field surveys- associational							
20	Calcifuge grasslands.	Sixty four sites (from southwest England to northern Scotland) were selected from the UK National Vegetation Classification of U4 ‘*Festuca ovinae-Agrostis capillaries-Galium saxatile* grasslands’, which represent calcifuge	Occurring components included: *Festuca rubra*, *Pseudoscleropodium purum*, *Hylocomium splendens*, *Euphrasia officinalis*, *Plantago lanceolata*, *Viola riviniana*, *Rhytidiadelphus triquetrus*, *Luzula multiflora*, *Succisa pratensis*, *Vaccinium vitis-idaea*, *Lathyrus montanus*, *Campanula rotundifolia*, *Anthoxanthum odoratum*, *Deschampsia flexuosa*, *Nardus stricta*, *Hypnum cupressiforme* agg., *Dicranum scoparium*.	1 g.s.	AOT40 was used as an index of O_3 exposure.	Even the sites were not selected according to O_3 gradient species response displayed a decline in O_3-susceptible species and an increase in O_3-tolerant plant species with increasing O_3 exposure. This study suggested that O_3 had the highest impact on community composition of acid grasslands (at national level) but has no significant impact on diversity itself, evaluated either by species richness or by diversity indices	(*53*)

		grasslands widely distributed in the British Isles.				incorporating equitability (Shannon’s H or Simpson’s D).	

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