

Impacts of Elevated Atmospheric CO₂ and O₃ on Forests: Phytochemistry, Trophic Interactions, and Ecosystem Dynamics

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Abstract Prominent among the many factors now affecting the sustainability of forest ecosystems are anthropogenically-generated carbon dioxide (CO₂) and ozone (O₃). CO₂ is the substrate for photosynthesis and thus can accelerate tree growth, whereas O₃ is a highly reactive oxygen species and interferes with basic physiological functions. This review summarizes the impacts of CO₂ and O₃ on tree chemical composition and highlights the consequences thereof for trophic interactions and ecosystem dynamics. CO₂ and O₃ influence phytochemical composition by altering substrate availability and biochemical/physiological processes such as photosynthesis and defense signaling pathways. Growth of trees under enriched CO₂ generally leads to an increase in the C/N ratio, due to a decline in foliar nitrogen and concomitant increases in carbohydrates and phenolics. Terpenoid levels generally are not affected by atmospheric CO₂ concentration. O₃ triggers up-regulation of antioxidant defense pathways, leading to the production of simple phenolics and flavonoids (more so in angiosperms than gymnosperms). Tannins levels generally are unaffected, while terpenoids exhibit variable responses. In combination, CO₂ and O₃ exert both additive and interactive effects on tree chemical composition. CO₂- and O₃-mediated changes in plant chemistry influence host selection, individual performance (development, growth, reproduction), and population densities of herbivores (primarily phytophagous insects) and soil invertebrates. These changes can effect shifts in the amount and temporal pattern of forest canopy damage and organic substrate deposition. Decomposition rates of leaf

litter produced under elevated CO₂ and O₃ may or may not be altered, and can respond to both the independent and interactive effects of the pollutants. Overall, however, CO₂ and O₃ effects on decomposition will be influenced more by their impacts on the quantity, rather than quality, of litter produced. A prominent theme to emerge from this and related reviews is that the effects of elevated CO₂ and O₃ on plant chemistry and ecological interactions are highly context- and species-specific, thus frustrating attempts to identify general, global patterns. Many of the interactions that govern above- and below-ground community and ecosystem processes are chemically mediated, ultimately influencing terrestrial carbon sequestration and feeding back to influence atmospheric composition. Thus, the discipline of chemical ecology is fundamentally important for elucidating the impacts of humans on the health and sustainability of forest ecosystems. Future research should seek to increase the diversity of natural products, species, and biomes studied; incorporate long-term, multi-factor experiments; and employ a comprehensive “genes to ecosystems” perspective that couples genetic/genomic tools with the approaches of evolutionary and ecosystem ecology.

Keywords CO₂ · Decomposition · Ecosystem processes · Forest · Global change · Herbivory · Human impacts · Nutrient cycling · Ozone · Phenolics · Phytochemistry · Plant-insect interactions · Tannins · Terpenoids

Introduction

Forest ecosystems have played roles of unparalleled importance in the evolution and ecology of life on Earth. Currently, forests cover ~30% of the earth's land surface, store ~45% of terrestrial carbon, and contribute ~50% of

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terrestrial net primary production (Bonan 2008). They provide vital ecological, economic and social services, including regulation of biogeochemical cycles, production of food and fiber, preservation of biodiversity, governing of climate systems, and buffering against natural and anthropogenic disasters (Hassan et al. 2005; Millenium Ecosystem Assessment 2005).

Earth's forests are now subject to a veritable Pandora's Box of environmental evils, the magnitude and diversity of which have never occurred previously in human history. Worldwide, climate and land use changes, invasive species, pest outbreaks, and pollution are exacting an increasing toll on the health, biological diversity, and sustainability of forests (Laurence and Peres 2006; Hari and Kulmala 2008; Kurz et al. 2008; Raffa et al. 2008; van Mantgem et al. 2009). Against this backdrop of massive and seemingly intractably complex socio-environmental ills, the question arises: "of what relevance is chemical ecology?"

In a word, plenty. Naturally-produced chemicals, particularly phytochemicals, govern many, if not most, of the interactions that characterize community and ecosystem function. They *respond* to global environmental change; they *perpetuate*, via interaction networks, the consequences of global change; and they *feed back* to influence future global change. Consider, for example, the carbon cycle (Fig. 1). Enriched atmospheric CO₂ influences plant physiology, with consequences for plant production and chemical composition. Plant chemical composition in turn influences trophic interactions and decomposition, which ultimately feed back to affect atmospheric CO₂ concentrations (Peñuelas and Estiarte 1998; Beedlow et al. 2004; Lindroth and Dearing 2005). Or, consider the isoprenoids and related biogenic volatile organic carbons (BVOCs) released in large quantities by some species of trees (e.g., eucalypts, poplars, and conifers). These compounds are precursors for the formation of tropospheric ozone (O₃), which in turn negatively impacts plant production (Fig. 1) and human health (Fowler et al. 1999; Lerdau and Gray 2003; Monson 2003; Laothawornkitkul et al. 2009). An understanding of the roles of natural products as responders to, and effectors of, global change will improve our ability to predict ecosystem responses to future change, and inform management decisions with respect to mitigation and adaptation strategies.

Analogous in some respects to intestinal villi, forest trees provide structurally expansive and heterogeneous surfaces through which the exchange of chemicals between the atmosphere and biosphere is effected. The same properties enhance exposure of forest trees to gaseous pollutants. Two such gases, CO₂ and O₃, are considered the most important and ubiquitous, affecting forests worldwide (Saxe et al. 1998; Fowler et al. 1999; Felzer et al. 2004; Karnosky et al. 2005, 2007). Independent of their function as greenhouse

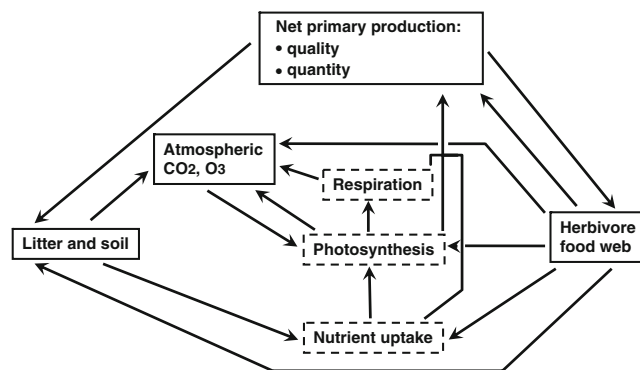


Fig. 1 Carbon cycling and storage in forest ecosystems. Solid boxes represent major pools; dashed boxes represent major plant physiological processes (adapted from Lindroth and Dearing 2005)

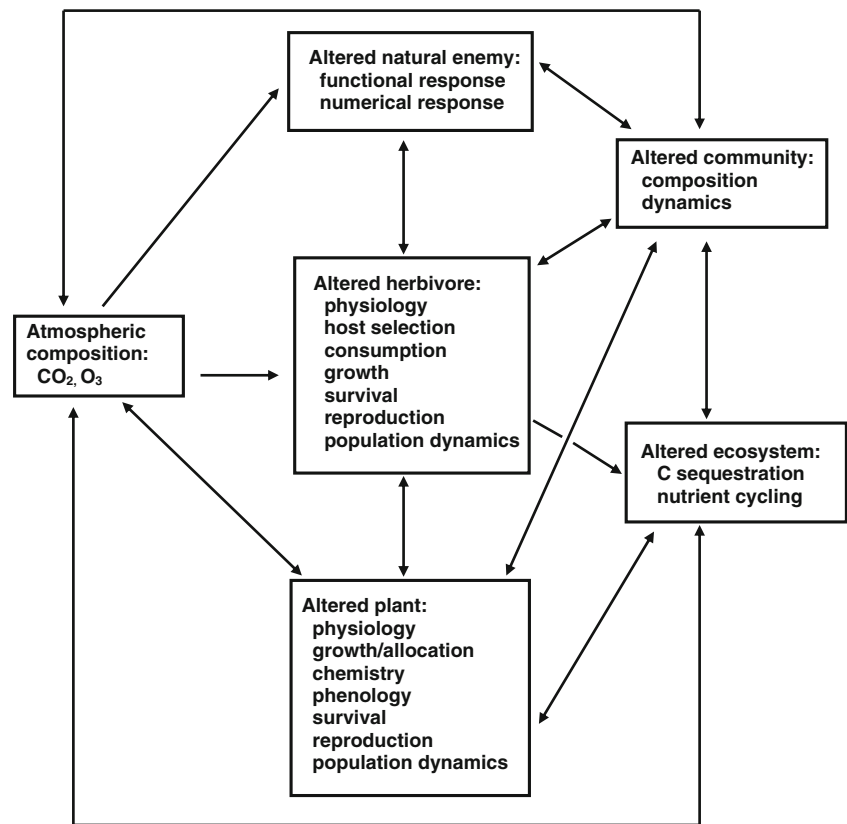
gases, CO₂ and O₃ are directly and indirectly affecting forest ecosystems now, with increasingly pronounced impacts predicted for the future (Fig. 2). (Note that although both CO₂ and O₃ occur naturally, they are appropriately considered "pollutants" because of their anthropogenic production and effects on living systems.)

The purpose of this review is to assess the impacts of enriched atmospheric CO₂ and O₃, independently and interactively, on the chemical ecology of forest ecosystems. Several recent reviews provide an assessment of the effects of CO₂ and O₃ on plant chemistry; that information will be summarized and updated here. I will then explore the consequences of CO₂- and O₃-mediated changes in tree chemistry for trophic interactions (plant—animal—natural enemy) and ecosystem dynamics (rates of plant damage, organic substrate deposition and decomposition) (Fig. 1). I will not detail the links between ozone and plant volatiles, as those have been covered in several very recent reviews, including elsewhere in this issue (Laothawornkitkul et al. 2009; Pinto et al. 2010; Yuan et al. 2009). Finally, although CO₂ and O₃ are both greenhouse gases, the effects of climate change per se on ecological interactions (e.g., Bale et al. 2002; Parmesan 2006; Berggren et al. 2009; Dukes et al. 2009; Pelini et al. 2009) are beyond the scope of this review.

Temporal Trends in Concentrations of CO₂ and O₃

Concentrations of atmospheric CO₂ now approach 386 ppm, higher than at any time in the last 26 million years (Pearson and Palmer 2000) and 40% higher than at the dawn of the industrial revolution. Emissions scenarios predict a range of substantially elevated concentrations by the end of this century, from a low of 550 ppm to a high of 900 ppm (Karl et al. 2009). Over the last several decades, combustion of fossil fuels, coupled with a smaller contribution from manufacture of cement, have accounted for

Fig. 2 Complex of interacting factors that feed forward and feed back to influence food webs, community and ecosystem structure and function, and atmospheric composition



approximately 80% of human-caused emissions. The remaining 20% has resulted from changes in land use, primarily deforestation (Denman et al. 2007; Karl et al. 2009).

Since the evolution and spread of large, vascular plants, about 380 MYA, forests have played prominent, regulating roles with respect to atmospheric CO₂ concentrations. They were largely responsible for the massive drawdown of atmospheric CO₂ (peaking at upwards of 5,000 ppm) in the late Devonian and Carboniferous Periods. Forests exerted two major effects on the global carbon cycle, both of which reduced atmospheric CO₂ concentrations (Berner 2005). First, they produced massive amounts of lignified tissue that was refractory to decomposition. This organic material, upon sedimentary burial, led to the formation of enormous coal deposits. Second, large and deep root systems greatly accelerated nutrient uptake from rock substrates, enhancing the weathering of Ca-Mg silicate minerals. Ca and Mg cations, in the presence of carbonic acid (derived from atmospheric CO₂ and water), formed Ca-Mg carbonates, which precipitated in ocean sediments to form limestone and dolomite.

Tropospheric ozone is recognized as the most damaging and widespread pollutant affecting agricultural and forest ecosystems in North America and Europe (Chameides et al. 1994; Ollinger et al. 1997; Skärby et al. 1998; Fuhrer and Booker 2003; Ashmore 2005; Karnosky et al. 2007; Wittig

et al. 2009). Concentrations of O₃ have increased an average of 38% (range of 20–50%) since the pre-industrial age (Denman et al. 2007). Although O₃ is a regional pollutant, large areas of the earth already are impacted, background levels are steadily increasing, and 60% of the world's forests are expected to be affected detrimentally by the end of this century (Fowler et al. 1999; Vingarzan 2004; Denman et al. 2007).

O₃ is a secondary pollutant, produced via the catalytic oxidation of hydrocarbons (e.g., VOCs) by OH radicals, in the presence of nitrogen oxides and sunlight (Wennberg and Dabdub 2008). Because O₃ is highly reactive, it persists for short periods (hours to several weeks) in the atmosphere. Rapid degradation, coupled with substantial spatial and temporal variation in production, contribute to highly variable distribution of this pollutant. Regional VOC and NO_x emission control policies are expected to reduce peak O₃ levels in North American and Europe, whereas in Asia and other regions, emissions of these precursors are increasing (Ashmore 2005). Moreover, background levels of O₃ (not attributable to local origin) are increasing steadily worldwide (Vingarzan 2004).

Plant-derived, biogenic VOCs can be important contributors to the formation of O₃ (Sharkey and Lerdau 1999; Lerdau and Gray 2003; Laothawornkitkul et al. 2009; Yuan et al. 2009). When BVOCs such as isoprene, monoterpenes or methylbutenol are oxidized in the presence of NO_x,

substantial amounts of O₃ can be produced. Indeed, large inputs of BVOCs may handicap efforts to reduce ozone pollution through controlling emissions of anthropogenic VOCs. Moreover, ecological interactions may amplify ozone production. A modeling study by Litvak et al. (1999) indicated that even modest (10%) damage by defoliating insects in coniferous forests is sufficient to increase local concentrations of O₃.

In contrast, in the presence of low NO_x concentrations, BVOCs are oxidized by O₃, leading to a decrease in atmospheric O₃ and a concomitant increase in secondary organic aerosols (Laothawornkitkul et al. 2009). Thus, forest ecosystems can serve as effective quenchers of anthropogenic O₃, but in the process contribute to atmospheric aerosol loading that influences climate. Kurpius and Goldstein (2003) documented that for a *Pinus ponderosa* forest during summer, 51% of atmospheric O₃ is broken down via gas phase chemistry (reactions with BVOCs), 30% is lost to stomatal uptake, and 19% is lost to surface deposition.

Effects of CO₂ and O₃ on Plant Physiology and Growth

Carbon Dioxide

As the substrate for photosynthesis, and ultimate source of all carbon fixed via primary production, atmospheric CO₂ strongly influences plant physiology and growth. Photosynthesis is catalyzed by Rubisco (ribulose-1,5-bisphosphate carboxylase oxygenase), which fixes carbon to form carbohydrates. In C₃ plants, including most forest tree species, enriched CO₂ atmospheres increase the intracellular CO₂:O₂ ratio, thus reducing photorespiration (oxygenation) and increasing photosynthesis (carboxylation). Carbohydrates then are transported throughout the plant and subsequently converted to various carbon-containing compounds required for plant metabolism, structure, storage and defense (Fig. 3).

The effects of elevated atmospheric CO₂ on tree physiology and growth have been covered in numerous reviews (e.g., McGuire et al. 1995; Saxe et al. 1998; Long et al. 2004; Ainsworth and Long 2005; Norby et al. 2005; Körner 2006; Leakey et al. 2009). In brief, N use efficiency improves as the N allocated to Rubisco (the largest single pool of N in leaves) can be re-allocated to other metabolic processes. Stomatal conductance generally declines, decreasing transpiration and improving water use efficiency. Enhanced photosynthesis, coupled with improved nitrogen and water use efficiencies, often accelerates plant growth, i.e., the “fertilizer effect” of enriched CO₂ (Beedlow et al. 2004). The growth stimulation afforded by enriched CO₂ is especially strong in young forest stands, characterized by abundant soil resources and open canopies. In contrast,

studies in mature stands, where tree growth is tightly coupled to soil nutrients and canopies are closed, indicate smaller to zero growth enhancement (Asshoff et al. 2006; Körner 2006).

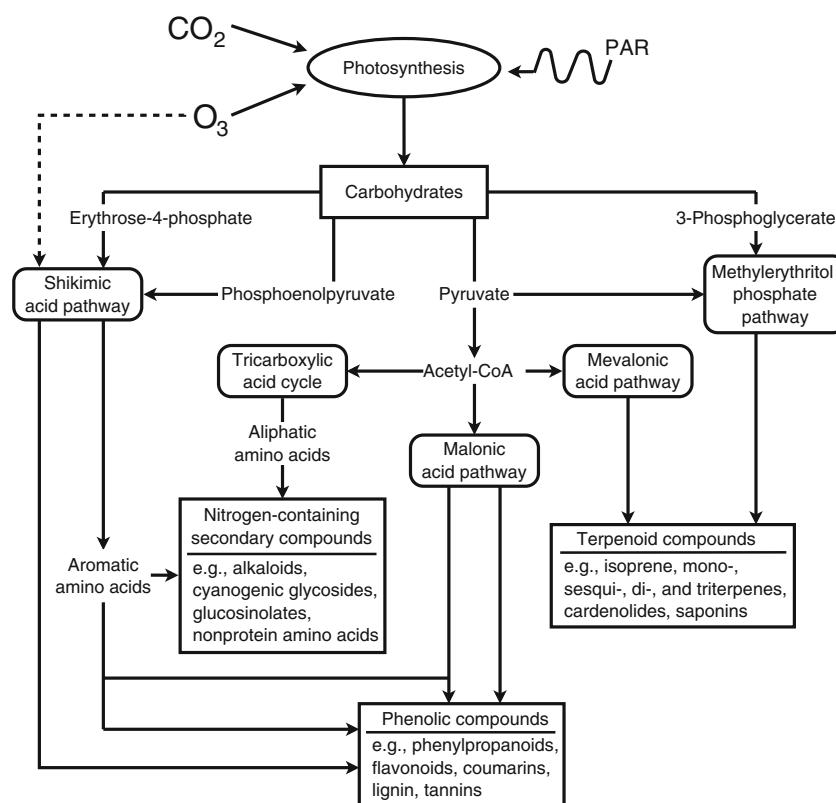
A key, unresolved issue in global change biology is to what extent improved tree growth can be sustained into the future, affording greater carbon sequestration and thereby mitigating climate change. To date, open-air FACE studies generally indicate that accelerated rates of photosynthesis and growth are sustained over the short to medium term and across a broad range of productivity (Long et al. 2004; Norby et al. 2005; Huang et al. 2007; but see Körner et al. 2005 for a contrary example with mature trees). Still, the duration of these studies is short compared with the lifespan of trees. The Progressive Nitrogen Limitation (PNL) hypothesis (Luo et al. 2004) posits that CO₂-enhanced growth and carbon sequestration will diminish over time: as storage of N in plant biomass and soil organic matter increases, available soil N becomes limiting to growth. If not nitrogen, then a host of other factors (climate change, fire, insects, ozone, etc.) is predicted to constrain the fertilization effect of atmospheric CO₂ enrichment (Beedlow et al. 2004; Huang et al. 2007). For example, Cole et al. (2009) reported that atmospheric CO₂ enrichment over the last 50 years has increased growth rates of natural stands of *Populus tremuloides* by an astonishing 53%, but enhancement is diminished in periods of low precipitation. Finally, elevated atmospheric CO₂ may, via climate change impacts, ultimately lead to reduced carbon sequestration at the landscape level. In Canada, for example, extensive areas of forest recently have transitioned from CO₂ sinks to CO₂ sources, due to insect outbreaks and fires (Kurz et al. 2008).

Ozone

Reactions of tropospheric O₃ with plant chemistry can occur in gaseous, solid, or liquid phases (Fuhrer and Booker 2003). Gaseous reactions with plant VOCs are described by Pinto et al. (2010). Solid phase reactions may occur, for example, with plant cuticular components. The most important reactions for plants, however, occur in the liquid phase, after ozone has entered through the stomata and diffused into the aqueous media of intercellular spaces (Fuhrer and Booker 2003).

O₃ elicits a cascade of damaging physiological consequences in plants, described in detail in numerous recent reviews (e.g., Andersen 2003; Fuhrer and Booker 2003; Ashmore 2005; Kangasjärvi et al. 2005; Wittig et al. 2007, 2009). Because it is so highly reactive, O₃ is minimally distributed following dissolution into intercellular fluids. It reacts with lipid and protein components of cell walls and plasma membranes, leading to the formation of aldehydes, peroxides and assorted reactive oxygen species (ROS). These products

Fig. 3 Biosynthetic pathways for the production of the major classes of plant secondary compounds (adapted from Taiz and Zeiger 2002). Dashed lines indicate a control (signal transduction) pathway. PAR = photosynthetically active radiation



can then cause cell death, or activate various transduction pathways for defense responses, such as stomatal closure, production of anti-oxidants such as ascorbate and phenolics, and programmed cell death (a component of the hypersensitive response) (Fuhrer and Booker 2003; Valkama et al. 2007; Fig. 3). Reduced stomatal conductance, compromised photosynthesis, consumption of carbohydrates in localized tissue repair, and inhibition of phloem transport at the leaf level combine to decrease the supply of carbohydrates throughout the plant (Andersen 2003; Wittig et al. 2007, 2009). As a consequence, senescence is accelerated and whole-plant growth is inhibited.

A recent, comprehensive meta-analysis by Wittig et al. (2009) evaluated the effects of O₃ on the growth of trees representative of northern temperate and boreal forests. They found that, relative to preindustrial levels, current levels of tropospheric O₃ account for an average 7% reduction in tree biomass growth. Moreover, as O₃ levels continue to rise, further decreases of 11% and 17% are predicted for 2050 and 2100, respectively.

Understanding of the interactive effects of CO₂ and O₃ on tree growth is critically important to our ability to predict the efficacy of forests to serve as global carbon sinks in the future (Beedlow et al. 2004). A modeling study by Sitch et al. (2007) indicated that tropospheric O₃ may reduce terrestrial carbon sinks, and thereby effect indirect radiative forcing of climate change. This conclusion was subsequently corroborated by the work of Wittig

et al. (2009), which indicated increasingly pronounced reductions in tree growth under elevated concentrations of O₃ in the future. To date, only one experiment has addressed the long-term (10+ years) effects of independent and combined CO₂ and O₃ treatments on forest trees. Karnosky et al. (2005) reported that at Aspen FACE (Wisconsin, U.S.A.), O₃ reduced the growth-enhancement effect of CO₂, although responses differed among tree species.

Effects of CO₂ and O₃ on Phytochemistry

CO₂ and O₃ directly and indirectly influence carbon assimilation and nutrient (e.g., N) acquisition, and thereby the pools and fluxes of key precursors for the production of secondary metabolites (Fig. 3). They also may influence the signal transduction pathways that govern gene expression and subsequent synthesis of secondary compounds. Not surprisingly then, growth under CO₂- or O₃-enriched atmospheres typically alters the chemical composition of forest trees.

Numerous reviews have addressed the effects of CO₂ and O₃ on plant chemistry (Kangasjärvi et al. 1994; Peñuelas et al. 1997; Bezemer and Jones 1998; Cotrufo et al. 1998a; Koricheva et al. 1998a; Peñuelas and Estiarte 1998; Norby et al. 2001; Zvereva and Kozlov 2006; Stiling and Cornelissen 2007; Valkama et al. 2007; Bidart-Bouzat and Imeh-

Nathaniel 2008; Wittig et al. 2009). Unless noted otherwise, the general trends for forest trees, summarized below, are drawn from that comprehensive set of reviews. Specific studies will be mentioned to illustrate particular patterns or highlight recent findings.

To date, research on the effects of CO₂ and O₃ on forest trees has been restricted to a small set of primarily temperate species. Most commonly studied are species of *Acer*, *Betula*, *Fagus*, *Pinus*, *Populus*, and *Quercus*. Although these are prominent, if not “foundation,” genera, a major challenge to the global change research community is to broaden the taxonomic and geographic representation of trees, especially tropical species.

Carbon Dioxide

Enriched atmospheric CO₂ produces generally consistent effects on concentrations of primary metabolites in trees (Cotrufo et al. 1998a; Koricheva et al. 1998a; Saxe et al. 1998; Stiling and Cornelissen 2007). Foliar concentrations of nitrogen typically decline (on average, 14–17% relative to ambient CO₂), due to reductions in Rubisco concentrations and dilution by accumulating carbohydrates. Levels of simple sugars may or may not increase, whereas levels of starch generally increase substantially.

Studies of the effects of CO₂ on secondary metabolites in trees have been restricted to “carbon-based” compounds, particularly phenolics and terpenoids. Consequently, this research often has been framed in the context of source-sink balance hypotheses [e.g., carbon-nutrient balance hypothesis (Bryant et al. 1983), growth-differentiation balance hypothesis (Herms and Mattson 1992)]. These hypotheses predict that elevated CO₂ will increase plant concentrations of carbon-based secondary compounds, especially if soil nutrient availability is limiting to growth (Peñuelas and Estiarte 1998).

In general, results have not been as readily predictable as the hypotheses would suggest (reviewed in Koricheva et al. 1998a; Peñuelas and Estiarte 1998; Zvereva and Kozlov 2006; Stiling and Cornelissen 2007; Bidart-Bouzat and Imeh-Nathaniel 2008). Moreover, the shikimic acid pathway, leading to production of phenolics, is more strongly influenced by CO₂ levels than are the mevalonic acid and methylerythritol phosphate pathways, leading to production of terpenoids (Fig. 3). Enriched atmospheric CO₂ typically, but not invariably, increases tannin concentrations. Common, but less consistent, are increases in simple phenolics, including phenolic acids and glycosides (e.g., salicylates). Growth under elevated CO₂ also tends to increase lignin concentrations in tree leaves (Coûteaux et al. 1999; Norby et al. 2001), although more recent results, from long-term FACE studies, have shown no such effects (Finzi et al. 2001; Parsons et al. 2008; Liu et al. 2009). Overall, effects

of CO₂ on terpenoid concentrations are minimal; although a few studies have revealed increases, most have found no change or decreases in concentrations.

The C/N ratio is a widely recognized, general index of overall tissue quality, relevant to both herbivory and decomposition. Enriched CO₂ atmospheres almost invariably increase C/N ratios in tree foliage (Coûteaux et al. 1999), due to a combination of decreased N concentrations and increased carbohydrate and phenolic concentrations.

Aside from such general indices, however, empirical studies have frustrated attempts to develop general, predictive models of the impacts of CO₂ on plant secondary chemistry. In short, a host of interacting factors have, over evolutionary time, combined to determine the chemical profiles of trees, and the effects of any one environmental factor can be masked by the direct or interactive effects of others.

One of the major confounding factors is taxon-specificity. Phytochemical responses of trees to CO₂ enrichment differ at the level of class (e.g., gymnosperm vs. angiosperm), species, and genotype (Julkunen-Tiitto et al. 1993; Lindroth et al. 2001, 2002; Zvereva and Kozlov 2006; Stiling and Cornelissen 2007; Bidart-Bouzat and Imeh-Nathaniel 2008). These differences represent taxonomic variation in evolutionary strategies that optimize solutions for the conflicting demands of carbon allocation toward growth, reproduction, and defense. For example, Cseke et al. (2009) recently compared leaf transcription profiles, physiology, and biochemistry between CO₂-responsive and unresponsive clones of *Populus tremuloides* grown at Aspen FACE. They found that the CO₂-responsive clone partitions carbon into pathways associated with “active” defense and stress responses, carbohydrate synthesis, and subsequent growth, whereas the unresponsive clone partitions carbon into pathways associated with cell wall compounds such as lignin, derived from the shikimic acid pathway.

Interactions with abiotic environmental factors also influence chemical responses of trees to CO₂. The availability of key resources (e.g., nutrients, water, light) can strongly shape plant chemical responses to CO₂, but, again, the magnitude and even direction of responses differ among chemical constituents and tree species (e.g., Lavola and Julkunen-Tiitto 1994; Kinney et al. 1997; McDonald et al. 1999; Booker and Maier 2001; Coley et al. 2002; Koike et al. 2006). Of particular importance in the context of global environmental change are potential interactions between atmospheric CO₂ and temperature. To that end, Zvereva and Kozlov (2006) conducted a meta-analysis of studies that assessed the simultaneous elevation of CO₂ and temperature on plant quality. Again, results were compound- and taxon-specific. Thus, responses to enriched CO₂ may be: 1) independent of temperature (e.g., foliar nitrogen and phenolics in Angiosperms); 2) offset by elevated temperature

(e.g., carbohydrates; foliar terpenes in gymnosperms); or 3) apparent only with elevated temperature (e.g., foliar nitrogen in gymnosperms, phenolics, and terpenoids in woody tissues). More recently, Veteli et al. (2007) reported for three tree species (*Betula* and *Salix*) that carbon allocation to phenolics was increased under elevated CO₂, decreased under elevated temperature, and unchanged under a combination of elevated CO₂ and temperature.

Interactions with biotic agents also may influence chemical responses of trees to CO₂, although much less research has been conducted with biotic than with abiotic factors. Herbivory frequently elicits induced chemical changes in trees, particularly in levels of phenolics (Nykänen and Koricheva 2004). Thus, enriched CO₂ atmospheres have been predicted to enhance induced chemical responses. Evidence to date, however, suggests that CO₂ does not modify induced chemical responses to defoliation in trees, although only a few studies have been conducted (Lindroth and Kinney 1998; Roth et al. 1998; Agrell et al. 1999; Rossi et al. 2004; Hall et al. 2005; Huttunen et al. 2008).

A final factor complicating attempts to draw general conclusions about the effects of CO₂ on tree chemistry is ontogeny (genetically-determined developmental changes). Many species of trees exhibit strong ontogenetic shifts in chemical composition (e.g., Bryant and Julkunen-Tiitto 1995; Donaldson et al. 2006), which reflect shifting physiological demands and defense strategies as trees mature (Boege and Marquis 2005). How ontogeny may interact with CO₂ to affect chemical profiles in trees is unknown. Such interactions may be one reason, however, for why chemical responses of plants in long-term FACE studies appear to be less pronounced than in short-term pot studies (Ainsworth and Long 2005; Lindroth, unpublished data).

Ozone

Elevated levels of tropospheric O₃ can modify plant chemical composition via several mechanisms. By disrupting photosynthesis, O₃ inhibits production of carbohydrates and the flow of precursors into pathways of primary and secondary metabolism (Fig. 3). Ozone also functions as a general abiotic elicitor of defense signaling pathways, particularly of phenolic compounds (Kangasjärvi et al. 1994; Heath 2008; Betz et al. 2009a). The two mechanisms may, of course, interact, as reductions in carbohydrate precursors for the shikimic acid pathway may constrain induced defensive responses. Moreover, these mechanisms may play out differently over different time scales, resulting in varying responses to acute versus chronic ozone stress.

The effects of O₃ on levels of primary metabolites vary among compounds and tree species. Levels of nutrients

(e.g., N, P, K, Ca) may increase, decrease, or not change in response to O₃ fumigation. Levels of simple sugars generally are unaffected, whereas levels of starch typically decline in angiosperms, but not gymnosperms.

As is true for CO₂, studies of the effects of O₃ on secondary compounds in trees have been restricted almost entirely to phenolics and terpenoids. In general, elevated O₃ leads to increases in concentrations of phenolic acids and flavonoids in angiosperms but not in gymnosperms, and no overall change in concentrations of tannins. Foliar lignin concentrations also are generally unresponsive to O₃ fumigation (Boerner and Rebbeck 1995; Booker et al. 1996; Liu et al. 2005; Oksanen et al. 2005). Effects of O₃ on terpenoid levels have been studied in only a few tree species, and results are species-specific: increases in *Pinus sylvestris* and *Populus* species, and no change in *Picea abies* (Valkama et al. 2007; Blande et al. 2007). Moreover, the magnitude of response varies among types of terpenes, with diterpenes responding more strongly to O₃ than mono- or sesquiterpenes.

That O₃ elicits increases in the levels of some carbon-based secondary metabolites without corresponding increases in carbohydrates runs counter to the predictions of source-sink balance hypotheses (e.g., Bryant et al. 1983), and may be explained by the fact that ozone triggers up-regulation of antioxidant defense systems linked to the shikimic acid pathway (Valkama et al. 2007). Ozone causes oxidative damage, coupled to the proliferation of oxygen radicals. Gene transcription and activity of numerous enzymes in the shikimic acid pathway are elevated in response to O₃ exposure, leading to the production of antioxidants such as flavonoids and other simple phenolics, and shifts in the monomeric composition of lignin (Kangasjärvi et al. 1994; Heath 2008; Betz et al. 2009a, b). Indeed, ozone-induced phenolics have been considered potential bioindicators of O₃ stress in plants under natural conditions (Sager et al. 2005; Bidart-Bouzat and Imeh-Nathaniel 2008).

Ozone exposure also influences the amount and chemical composition (e.g., alkyl esters and fatty acids vs. alkanes and alkanols) of cuticular waxes in tree leaves (Karnosky et al. 2002; Kontunen-Soppela et al. 2007; Percy et al. 2009). These changes in turn determine the physical structure of the cuticular surface, with repercussions for leaf surface properties such as wettability.

In contrast to a large body of research with CO₂, little work has addressed how O₃ pollution interacts with other environmental factors to shape plant chemical profiles. Lindroth et al. (1993a) evaluated the effects of elevated O₃ and light intensity on several foliar constituents of hybrid poplar and sugar maple (*Acer saccharum*), and found that O₃ and light interacted to affect only concentrations of N in maple. Given that forest ecosystems are exposed to

multiple, simultaneous stress factors (e.g., climate change, insect outbreaks) in addition to increasing O₃ damage, and that some of those stressors induce the same chemical defense pathway as does O₃, such research is sorely needed.

Interactions Between CO₂ and O₃

The environmental factor that has been investigated most thoroughly in combination with O₃ pollution is CO₂. Enriched CO₂ may reduce ozone damage to trees both by decreasing stomatal conductance (and consequent oxidative stress) and by increasing carbohydrate pools for the synthesis of antioxidant compounds. The recent meta-analysis by Valkama et al. (2007) evaluated the effects of O₃, and O₃ combined with CO₂, on foliar chemistry of 22 species of trees. They found that enriched CO₂ can offset O₃-induced chemical changes, but their analysis did not differentiate between simple additive, versus interactive, effects of CO₂ and O₃.

Indeed, CO₂ and O₃ exert both independent (additive) and interactive effects on tree chemical profiles. Elevated O₃ concentrations exacerbated the CO₂-mediated reductions in N concentrations in *Betula papyrifera* (Kopper et al. 2001) and *Populus tremuloides* (Holton et al. 2003), but offset reductions in N in *Betula pendula* (significant CO₂ × O₃ interactions). Peltonen et al. (2005) assessed the effects of CO₂ and O₃ on over thirty simple phenolics in *B. pendula*. They identified interactive effects in approximately three-quarters, and independent effects in one-quarter, of the cases. The interactive effects revealed that O₃-mediated induction of phenolics disappeared under elevated CO₂. Tannin levels also are influenced by both additive and interactive effects of CO₂ and O₃. Condensed tannin concentrations responded to the additive, independent effects of CO₂ and O₃ in *Fagus crenata* and various species of *Betula* (Kopper et al. 2001; Peltonen et al. 2005; Karonen et al. 2006), but to the interactive effects of CO₂ and O₃ in *Populus tremuloides* (Holton et al. 2003). Similarly, the effects of CO₂ and O₃ on monoterpenes and sesquiterpenes in *Pinus sylvestris* sometimes were additive and sometimes interactive (Sallas et al. 2001). In short, whether CO₂ and O₃ function in an additive or interactive manner depends on the particular chemical constituent and tree species studied.

Effects of CO₂ and O₃ on Trophic Interactions

Plant chemical composition is a strong ecological and evolutionary driver of trophic interactions, particularly between plants and herbivores. Given that green plants and phytophagous insects comprise nearly half of Earth's

recognized species (Strong et al. 1984), it is not surprising that these taxa occupy the vast majority of all studies on plant-herbivore interactions. Several recent reviews have addressed the effects of CO₂ and O₃ on plant-insect interactions (Zvereva and Kozlov 2006; Stiling and Cornelissen 2007; Valkama et al. 2007; Bidart-Bouzat and Imeh-Nathaniel 2008). Composite results will be summarized below and supplemented with additional, relevant studies of tree-feeding herbivores.

Carbon Dioxide

Little evidence exists to suggest that elevated CO₂, upwards of 1,000 ppm predicted for this century, will directly affect insects (Coviella and Trumble 1999). Work by Stange (1997) showed that atmospheric CO₂ concentrations influence host location by *Cactoblastis cactorum*, but no similar work has been done with forest insects. Rather, the major effect of enriched CO₂ on herbivores will be indirect, mediated via changes in plant quality (Fig. 2). Overall, elevated CO₂ reduces the quality of tree foliage as food, due to decreases in nitrogen (an index of protein) and minerals, and increases in carbohydrates and phenolics. These changes typically, but not always, alter the preference and reduce the performance of herbivores.

CO₂ concentration influences selection of plants by insects and mammals. Elevated CO₂ reduced leafminer oviposition on *Populus tremuloides* (Kopper and Lindroth 2003a), but increased birch aphid oviposition on one of two clones of *B. pendula* (Peltonen et al. 2006). The few feeding studies conducted to date suggest that preferences of insects for ambient versus elevated CO₂ foliage are not easily predicted, as shifts in both directions, and no change, have been reported (Traw et al. 1996; Kuokkanen et al. 2003; Agrell et al. 2005; Knepp et al. 2007). Elevated CO₂ decreased the palatability of winter-dormant *Betula* species to hares and rabbits (Mattson et al. 2004), but not to voles (Kuokkanen et al. 2004). Of greater relevance in the universally CO₂-enriched world of the future, however, is whether enrichment alters relative preferences of herbivores for various host species. Minimal evidence to date suggests that such is the case. For example, Agrell et al. (2005) demonstrated that forest tent caterpillars switch relative preferences between *Populus tremuloides* and *Betula papyrifera*, as well as between *P. tremuloides* genotypes, grown in high CO₂ environments.

Elevated CO₂ also influences the individual performance of herbivores, although the effects are highly species-specific for both trees and insects. Among the more uniform (though still variable) of responses is food consumption. In general, insects increase both consumption rates, and total consumption, on CO₂-enriched foliage, ostensibly as a means to compensate for reduced N concentrations. Compensatory

feeding responses may be constrained, however, by the simultaneous consumption of higher concentrations of secondary metabolites such as phenolics. Approximate digestibility of food typically is not significantly affected, but conversion efficiencies of digested food into biomass generally decline. Little is known about how CO₂ environment affects insect detoxication capacities; a study with gypsy moths revealed enhanced oxidase, reductase, and esterase activities in larvae fed CO₂-enriched *P. tremuloides*, but not *Acer saccharum* (Lindroth et al. 1993b). Development times usually are prolonged, while growth rates and final pupal and adult weights decline or are unaffected, but rarely improve. Effects on reproduction have rarely been investigated. Lindroth et al. (1997) found no effect of CO₂ on the number and mass of eggs produced by gypsy moths, whereas Awmack et al. (2004) reported decreased fecundity in aphids on *B. papyrifera* under elevated CO₂.

Species-specificity in terms of insect herbivore responses to enriched CO₂ may be linked to differences among feeding guilds (e.g., chewers, miners, sap-feeders, seed-feeders). Bezemer and Jones (1998) identified such guild effects for plants and phytophagous insects in general. Too few studies have been conducted with tree-feeding insects to evaluate differences among guilds. However, the minimal, if not positive effects of CO₂ on tree-feeding aphids (Docherty et al. 1997; Awmack et al. 2004), in contrast to effects on chewing insects, suggest that guild-specific responses to CO₂ may exist for forest insects.

Just as interactions with various environmental factors influence the effects of CO₂ on plant chemistry, so may they modulate effects on herbivores. For example, the effects of elevated CO₂ on herbivore performance can be ameliorated or exacerbated by the availability of light, water, and soil nutrients (e.g., Kinney et al. 1997; Lawler et al. 1997; Roth et al. 1997; Hättenschwiler and Schafellner 1999; Agrell et al. 2000). Potential interactions with climate warming are of particular interest, as warm temperatures generally affect insects in a manner opposite that of CO₂, i.e., accelerated development and enhanced growth and reproductive performance (Bale et al. 2002). Indeed, Zvereva and Kozlov (2006) conclude from their recent meta-analysis that temperature increases are likely to mitigate predicted negative effects of enriched CO₂ on insects, and emphasize the need for future research to address the two factors in parallel.

In addition to the bottom-up effects of altered plant chemistry, elevated CO₂ may influence the top-down effects of natural enemies (predators, parasitoids, and pathogens) on insect herbivores (Fig. 2). Enriched CO₂ could affect natural enemies by altering the behavioral or physiological defenses of prey, modifying the quality of prey (e.g., insect size, concentrations of plant-derived toxins), or by altering the efficacy of host location and disease transmission. Few studies have addressed these

possibilities, however, especially with forest systems. Elevated atmospheric CO₂ dramatically diminished the pheromone-mediated, predator escape behavior of aphids on *Populus tremuloides* (Mondor et al. 2004). In contrast, two studies have found the bottom-up effects of CO₂ on parasitoid performance (survivorship, development, and growth) to be minimal to nil (Roth and Lindroth 1995; Holton et al. 2003). The virulence of pathogens such as nuclearpolyhedrosis virus (NPV) is linked to host plant chemistry [e.g., tannins (Foster et al. 1992)], so CO₂-enriched atmospheres may influence the dynamics of disease outbreaks. CO₂-induced increases in condensed tannins in *P. tremuloides* did not, however, alter the susceptibility of gypsy moths to NPV (Lindroth et al. 1997). At this time insufficient information is available to predict whether the bottom-up effects of CO₂ on natural enemies will in general be buffered, amplified, or, more likely, context- and species-specific.

The impacts of elevated CO₂ concentrations on forest insects at the levels of populations and communities will be determined by a combination of bottom-up, top-down, and abiotic factors (Fig. 2). Historically, inferences about population-level responses have been drawn from studies of individual insects, but such conclusions can be misleading (Awmack et al. 2004). To date, however, few studies have addressed the impacts of elevated CO₂ at higher levels of organization. Several studies with tree-feeding aphids have shown no population-level effects (Docherty et al. 1997; Awmack et al. 2004), while another revealed positive effects of enriched CO₂ on aphid population size (Percy et al. 2002). In contrast, Stiling et al. (1999, 2002, 2003, 2009) found reduced densities (number per leaf) of herbivores, especially leafminers and leaf-tiers, in several woody species of a scrub-oak community exposed to elevated CO₂. Reduced herbivore densities were attributed to higher rates of both plant- and parasitoid-induced mortality, and were offset in several *Quercus* species by increased foliar production under high CO₂ (Stiling et al. 2009). Monitoring of aspen blotch leafminers (*Phyllonorycter tremuloidiella*) at Aspen-FACE indicated that elevated CO₂ roughly doubled plant-mediated mortality, substantially reduced predator-mediated mortality, and minimally influenced parasitoid-mediated mortality (Fig. 4).

Even fewer studies have addressed the effects of enriched CO₂ on speciose insect communities. Following 4 years of pan-trapping at Aspen FACE, Hillstrom and Lindroth (2008) reported that elevated CO₂ reduced abundance of phloem-feeding herbivores and increased abundance of chewing herbivores, although results were only marginally significant. Enriched CO₂ increased numbers of figitid and ichneumonid parasitoids, but not of braconid or chalcid parasitoids. Insect community composition differed between ambient and elevated CO₂ plots in 3 out of 4 years. In the most comprehensive study to

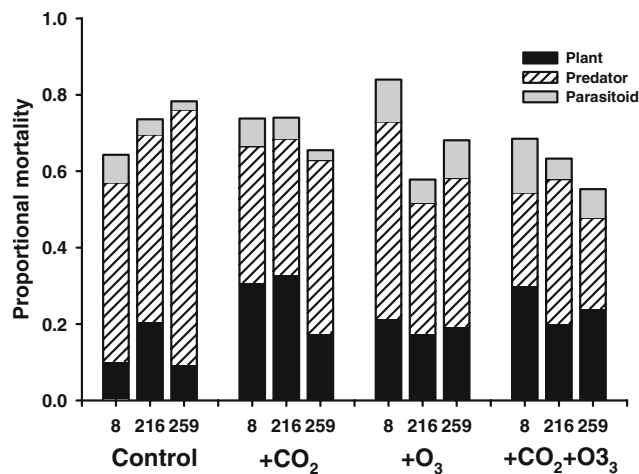


Fig. 4 Effects of CO₂ and O₃, in isolation and combination, on plant-, predator-, and parasitoid-induced mortality of aspen blotch leafminers (*Phyllonorycter tremuloidella*). Data reveal major sources of mortality, prior to adult emergence, for miners on *Populus tremuloides*. Numerals below bars indicate aspen genotypes. Observations recorded in July 2001 at the Aspen FACE research site (Awmack and Lindroth, unpublished data)

date for forest canopy insects, Hillstrom and Lindroth (unpublished data) visually monitored insects on *Populus tremuloides* and *Betula papyrifera* over 3 years at Aspen FACE, cataloging over 36,000 insects from nearly 300 species. Overall, enriched CO₂ tended to increase the abundance of phloem-feeders and decrease the abundance of chewers and gallers on *P. tremuloides*, although effects were variable among species and over time. Few consistent effects on abundance were found, however, for insects on *B. papyrifera*. Elevated CO₂ did not affect species diversity of canopy insects, except for insects on *P. tremuloides* in 1 year. Elevated CO₂ also did not influence insect community composition, except for insects on *B. papyrifera* in 1 year.

Ozone

Increasing evidence suggests that tropospheric ozone pollution may affect directly insects. Gate et al. (1995) found that under elevated O₃, searching efficiency of a hymenopteran parasitoid, as well as the proportion of hosts parasitized, declined. O₃ likely interacts with the complex of BVOCs that function as “infochemicals” to govern key interactions between insects and both their hosts (plant or insect) and natural enemies (predators and parasitoids) (Laothawornkitkul et al. 2009; Yuan et al. 2009; Pinto et al. 2010, this volume). Chemical degradation or transformation of BVOCs by O₃ is likely to increasingly disrupt olfactory cues in atmospheres of the future.

Selection of plants as oviposition or food hosts is influenced by O₃ concentrations. O₃-treated foliage reduced

leaf beetle oviposition onto *Populus deltoides* (Jones and Coleman 1988) and leafminer oviposition onto *Populus tremuloides* (Kopper and Lindroth 2003a). Few feeding studies have compared the preferences of insects for fumigated vs. nonfumigated foliage, and those indicate a range of responses. Leaf beetles preferred O₃-treated *P. deltoides* (Jones and Coleman 1988) and common leaf weevils preferred O₃-treated hybrid aspen (Freiwald et al. 2008), whereas forest tent caterpillars preferred untreated *P. tremuloides* (Agrell et al. 2005). Forest tent caterpillars greatly increased their preference for *Betula papyrifera*, relative to *P. tremuloides*, under elevated O₃, and also shifted their preferences among *P. tremuloides* genotypes (Agrell et al. 2005).

O₃ fumigation changes the individual performance of phytophagous insects, although, as for CO₂, effects are highly species-specific. The meta-analysis by Valkama et al. (2007) summarized results for 22 species of trees and ten species of insects. Food consumption rates typically are unaffected, as are survival and reproduction rates. Development times usually are reduced, and pupal masses increased, under elevated O₃. For example, Kopper and Lindroth (2003b) reported that for forest tent caterpillars reared on high-O₃ *Populus tremuloides*, development times decreased an average of 14% and pupal weights increased an average of 31% compared with insects on control trees. Relative growth rates of chewing, but not sucking, insects generally increase under elevated O₃. On average, insect fecundity is not affected by O₃, although Awmack et al. (2004) reported decreased fecundity for aphids on *Betula papyrifera*.

Under natural conditions, interactions between trees and herbivores likely are influenced by multiple, interacting stressors, including O₃ (Koricheva et al. 1998b). With the exception of interactions with CO₂ (addressed below), however, exceedingly little is known about how O₃ may interact with other environmental factors to influence trophic interactions. Lindroth et al. (1993a) reported that gypsy moth larval performance was influenced by the independent, but not interactive, effects of O₃ and light intensity. With the likelihood of increased pest damage to forests under future scenarios of global environmental change, assessments of the combined effects of ozone and other stressors should be a high priority.

A growing body of literature reveals that O₃ pollution likely will affect interactions between tree-feeding insects and their natural enemies. As indicated above and described elsewhere in detail (Yuan et al. 2009; Pinto et al. 2010), O₃ may directly affect the behavior and fitness of predators and parasitoids. Alternatively, elevated O₃ may alter the behavioral or physiological defenses of prey, or their quality as food. For example, Mondor et al. (2004) reported that at Aspen FACE, high O₃ environments markedly

enhanced predator escape behaviors of aphids on *Populus tremuloides*. Also at Aspen FACE, O₃ fumigation reduced survivorship of the parasitoid *Compsilura concinnata* (Holton et al. 2003). Decreased fitness was consistent with improved performance of its forest tent caterpillar host, which in turn was linked to decreased levels of phenolic glycosides in *P. tremuloides*.

Despite accumulating evidence for impacts of O₃ at the level of individual insect performance, little work has addressed impacts at the level of populations. Awmack et al. (2004) found that O₃ fumigation at Aspen FACE increased population densities of aphids on *Betula papyrifera* when protected from natural enemies, but not when unprotected. Also at Aspen FACE, elevated O₃ levels tended to increase plant-mediated mortality, reduce predator-mediated mortality, and increase parasitoid-mediated mortality of aspen blotch leafminers (*Phyllonorycter tremuloidiella*), although magnitudes of effects varied among insects on different aspen genotypes (Fig. 4).

At the level of insect communities, pan-trapping at Aspen FACE revealed positive effects of elevated O₃ on abundance of some phloem-feeding insects, and strong suppressive effects on a diversity of parasitoids (Hillstrom and Lindroth 2008). O₃ influenced the community composition of insects captured by traps in 2 out of 4 years. In our more recent visual censuses of canopy insects at Aspen FACE, we found that O₃ fumigation increased abundances of several species of chewing insects, but decreased numbers of several phloem-feeders, on *Populus tremuloides* (Hillstrom and Lindroth, unpublished data). O₃ did not, however, consistently affect abundances of insects on *Betula papyrifera*. O₃ also had generally little impact on the species diversity and community composition of canopy insects on several tree species, and across multiple years, at Aspen FACE.

Interactions Between CO₂ and O₃

Valkama et al. (2007) sought to differentiate the effects of O₃ alone, versus O₃ in combination with CO₂, on the performance of tree-feeding insects. Their meta-analysis revealed that many of the positive effects of O₃ on insect performance, especially for leaf chewers, are negated with simultaneous exposure to enriched CO₂. Their review did not, however, differentiate between simple additive, versus truly interactive, effects of CO₂ and O₃ on insects.

Indeed, relatively few studies have been designed to detect potential interactive effects of CO₂ and O₃ on forest insect performance, and those have shown mixed results. Several studies (Awmack et al. 2004; Peltonen et al. 2006) have assessed the independent and combined effects of CO₂ and O₃ on feeding, growth, and reproductive performance of aphids, and revealed no significant interactions. Similar-

ly, no significant interactions were observed with respect to CO₂ and O₃ effects on leafminers on *Populus tremuloides* (Kopper and Lindroth 2003a), whitemarked tussock moths feeding on *Betula papyrifera* (Kopper et al. 2001), or forest tent caterpillar feeding preferences for *P. tremuloides* and *B. papyrifera* (Agrell et al. 2005). In contrast, CO₂ and O₃ interacted to influence development and growth of forest tent caterpillars on *P. tremuloides*: O₃-mediated enhancements in performance were greater at ambient than elevated CO₂. In short, the limited evidence to date suggests that the combined effects of CO₂ and O₃ on insect herbivores will be primarily additive, although the standard caveat of context- and species-specificity applies.

Effects of CO₂ and O₃ on Ecosystem Dynamics

At the level of forest ecosystems, plant chemistry plays a host of important roles, the most prominent of which arguably is regulation of the transfer of fixed carbon to herbivores and decomposers—the dominant pathways of material flow (Fig. 1). Accumulating evidence suggests that elevated concentrations of CO₂ and O₃ will modify the complex of physiological and ecological interactions that determine rates of plant damage, organic substrate deposition, and nutrient cycling. In short, the effects of CO₂ and O₃ on plant chemistry will in turn influence the sequestration and cycling of carbon and nutrients in forest ecosystems.

Forest Canopy Damage

Phytophagous insects generally remove 2–15% of primary production in temperate deciduous forests (Cebrian 1999; Cyr and Pace 1993; Cebrian and Lartigue 2004), although during outbreaks, nearly 100% can be consumed (e.g., Donaldson and Lindroth 2008). Canopy damage rates are a function of the host preferences, individual consumption rates, population densities, and community composition of herbivorous insects, all of which can be influenced by levels of atmospheric CO₂ and O₃.

The effects of CO₂ on loss of primary production to herbivores have been explored in only a few forest systems. In a Florida scrub oak community, enriched CO₂ decreased the frequency of damage to *Quercus myrtifolia* (Stiling et al. 2002). Later work in the same system documented a decline in the frequency of damage by 4 of 6 insect feeding groups, across three *Quercus* species, under elevated CO₂ (Hall et al. 2005). Working with four understory tree species at the FACTS-1 FACE site (loblolly pine—hardwood forest), Hamilton et al. (2004) reported declines of 10–46% in foliar damage rates in high CO₂ plots. These declines were driven largely, however, by the

response of one species (*Ulmus alata*); damage rates in other species were not statistically significant from those of control trees. Knepp et al. (2005) followed up that work by censusing foliar damage rates in twelve species of potted saplings at FACTS-1 FACE. Enriched CO₂ reduced damage rates across all species in 2001, but not in 2002–2003. In the most comprehensive study to date, Couture, Meehan, and Lindroth (unpublished data) evaluated damage rates on leaves collected from *Populus tremuloides* and *Betula papyrifera* over 3 years at Aspen FACE. Elevated CO₂ concentrations did not markedly affect herbivory rates in 1 year, but increased damage rates by 2–3-fold in 2 years of the study. Overall, these studies indicate that canopy damage rates are likely to change under CO₂ concentrations of the future. But, consistent with effects on individual insects, the magnitude and direction of change will be both species-specific and temporally variable.

To date, no published study has evaluated the effects of elevated tropospheric O₃ on community-wide, canopy damage in forest ecosystems. Our work at Aspen FACE (Couture, Meehan and Lindroth, unpublished data) revealed that high levels of O₃ reduced damage to both *Populus tremuloides* and *Betula papyrifera* in 2 of 3 years.

Calculations of losses to primary production based on leaf areas damaged by herbivores may significantly underestimate true reductions in production. Aldea et al. (2006) showed that in a variety of deciduous tree species, chewing damage caused modest and localized suppression of photosynthetic efficiency in adjacent, undamaged leaf tissue, whereas galling damage caused large and extensive suppression. The authors suggest that the spatial propagation of reduced photosynthesis may be influenced by enriched CO₂, but did not observe such effects in their study. More recently, however, P. Nability and M. Hillstrom (pers. comm.) found that elevated CO₂ decreased the propagation of reduced photosynthesis caused by insects feeding on *Populus tremuloides* and *Betula papyrifera* at Aspen FACE.

Organic Substrate Deposition

Herbivorous insects mediate the transfer of organic materials from the forest canopy to the forest floor by depositing frass, greenfall, and insect biomass, and altering the timing and amounts of leachate throughfall and leaf litterfall. These processes have received little attention in the context of global change research.

We recently evaluated the effects of fumigation treatments at Aspen FACE on the quality of frass produced by whitemarked tussock moth larvae feeding on *Populus tremuloides*, and the effects of both frass and greenfall on soil microbial respiration and nitrogen leaching (Hillstrom, Meehan, Kelly and Lindroth, unpublished data). Insect frass

had slightly decreased N levels in high O₃ environments, and increased tannin levels in high CO₂ environments. However, neither frass nor greenfall quality affected microbial respiration (CO₂ efflux) or nitrogen (NO₃⁻) leaching in soil microcosms. In contrast, the quantity of frass and greenfall added to microcosms markedly affected both respiration and N leaching, under all atmospheric conditions. Thus, insects may affect carbon and nitrogen mineralization in forests of the future more through their effects on the quantity (and timing), than quality, of substrate deposition.

Indeed, atmospheric CO₂ and O₃ concentrations appear to influence rates of insect-mediated organic substrate deposition in forests. At Aspen FACE, rates of combined frass and greenfall deposition were 35% higher in elevated vs. ambient CO₂ stands, and 13% lower in elevated vs. ambient O₃ stands, over three field seasons (Meehan, Couture, Bennett and Lindroth, unpublished data).

Finally, insect feeding and atmospheric composition both may alter the timing of litterfall in forest ecosystems. In general, feeding damage accelerates leaf abscission, whereas enriched CO₂ may either accelerate (Stiling et al. 2002) or decelerate (Karnosky et al. 2003) abscission rates. How herbivory and atmospheric chemistry may interact to influence the magnitude and timing of leaf abscission is unknown.

Soil Invertebrates

Soil invertebrates play important roles in the nutrient cycling dynamics of forest ecosystems, from the comminution and microbial inoculation of plant litter to the vertical transport of organic matter through soil horizons. These processes are governed both by the quality and quantity of litterfall, which in turn are influenced by atmospheric composition (Fig. 1).

Several studies have evaluated the effects of leaf litter from elevated CO₂ plots on feeding preferences of isopods. Using short-term (3 day) feeding choice studies, Cotrufo et al. (1998b) found that isopods (*Oniscus asellus*) consumed 16% less high-CO₂ litter than ambient-CO₂ litter from *Fraxinus excelsior*. Subsequently, Cotrufo et al. (2005a) fed isopods (*Porcellio* sp.) for 2 weeks on litter from six tree species grown under ambient and elevated CO₂, and found no difference in palatability for five species, and increased preference for high-CO₂ litter for one species. Using no-choice feeding studies and two species of isopods, Hättenschwiler et al. (1999) found increased consumption of high-CO₂ beech litter, but no effect of CO₂ on consumption of spruce litter. Hättenschwiler and Bretscher (2001) reported that growth of trees under elevated CO₂ altered the relative preference of *O. asellus* for different litter species—increasing preference for one, decreasing

preference for another, and not affecting yet another. Thus, as is the case for insects feeding on green leaves, the consequences of CO₂ for detritus-feeding invertebrates are species-specific.

Additional studies have assessed the effects of leaf litter from elevated CO₂ or O₃ environments on fitness indices and population growth of soil invertebrates. Individual growth of juvenile earthworms (*Lumbricus terrestris*) was reduced when fed high-CO₂ or high-O₃ birch (*Betula pendula*) litter (Kasurinen et al. 2007), and high-CO₂ (but not high-O₃) aspen (*P. tremuloides*) litter (Meehan, Crossley and Lindroth, unpublished data). Growth and mortality rates of isopods, however, generally have been shown not to be affected by CO₂ or O₃ treatments (Hättenschwiler and Bretscher 2001; Kasurinen et al. 2007). Population growth of collembola (*Sinella curviseta*) decreased when reared on high-CO₂ aspen litter, but increased when reared on high-O₃ aspen litter (Meehan, Crossley and Lindroth, unpublished data).

Several studies have evaluated the effects of CO₂ and O₃ on soil invertebrate communities under field conditions. Hansen et al. (2001) investigated the effects of elevated CO₂ concentrations on soil microarthropods at the FACTS-1 FACE site. Over the first 19 months of CO₂ treatment, total microarthropod abundance declined by 34% in high-CO₂ plots, relative to ambient plots. The decline was driven by changes in abundance of oribatid mites, and was attributed to factors other than changes in litter quality. Haimi et al. (2005) censused populations of enchytraeids, mites and collembola in soil samples surrounding chambered *Pinus sylvestris* trees. Elevated CO₂ concentrations contributed to low numbers of acaridid mites, but did not significantly affect other soil fauna. Loranger et al. (2004) assessed responses of soil invertebrates to CO₂ and O₃ enrichment at Aspen FACE. They found that numbers of collembola and Oribatid mites, but not various other soil animals, declined in elevated CO₂ plots. Mites, but not other fauna, also decreased in high O₃ plots.

In summary, CO₂- and O₃-mediated changes in litter quality are likely to influence the performance of soil invertebrates of forests in the future. Some evidence exists to suggest that reduced litter quality will negatively affect the fitness of soil animals. In turn, their roles in nutrient cycling and soil carbon sequestration are likely to be altered. Overall, however, as noted by Coueteaux and Bolger (2000), no general patterns of response have yet emerged, likely due to the species- and context-specific nature of these interactions.

Litter Decomposition

Rates of litter decomposition and attendant nutrient mineralization are governed primarily by climate, litter chemistry,

and soil organisms. Climate (especially temperature and moisture) is the most important factor on a global scale, whereas chemical composition is most influential within a geographic region (Aerts 1997). All of these regulating factors are influenced directly, or indirectly, by CO₂ and O₃ (Fig. 1).

Litter quality is determined by carbon availability (which ranges from labile sugars to recalcitrant lignin), mineral content, and chemical modifiers such as tannins (Swift et al. 1979; Anderson 1991). Tannins and related phenolic constituents alter decomposition by complexing with other litter components (e.g., amino acids) and/or by inhibiting the degradative activity of enzymes and soil fauna (Horner et al. 1988; Anderson 1991; Coueteaux et al. 1995). For similar litter types in common environments, nitrogen, lignin, and tannin concentrations appear to act as “a hierarchical series of controls” (Anderson 1991). In high quality, readily decomposable litters, tannins may be rate retardants, whereas in low quality, slowly decomposable litters, lignin concentration, or lignin: N ratio typically determine decomposition rates (Anderson 1991). Any factor that markedly affects the nitrogen or C-based secondary metabolite concentration of litter also will affect decomposition, and consequently, nutrient availability (Chapin 1991).

The “litter quality” hypothesis, introduced by Strain and Bazzaz (1983), proposed that CO₂ enrichment will decrease plant nitrogen (relative to carbon) concentrations, and thereby reduce litter decomposition and soil N availability. Progressive soil nitrogen limitation is anticipated to eventually feed back to reduce plant production (Luo et al. 2004; Johnson 2006). Thus, CO₂-mediated changes in the chemical quality of litter have been of key interest with respect to understanding the cycling and storage of carbon in forest ecosystems. Indeed, litter from trees grown under elevated CO₂ characteristically exhibits reduced N concentrations and increased lignin concentrations and C:N ratios (Ceulemans et al. 1999; Norby et al. 2001).

To date, however, studies of the effects of elevated CO₂ on litter decomposition (measured as mass loss or microbial respiration) have not demonstrated a consistent direction of response. A comprehensive meta-analysis by Norby et al. (2001), including data for 33 woody species, showed that data do not support the “litter quality” hypothesis of reduced decomposition rates for litter produced under elevated CO₂. Decomposition studies published since that review have been increasingly realistic, employing more long-term and field-based approaches with open-top chambers or FACE sites. Still, a consistent pattern has yet to emerge. Several studies have documented reduced decomposition of leaf litter produced under elevated CO₂. For example, litter from three *Populus* species grown under elevated CO₂ at POPFACE exhibited reduced decomposi-

tion rates (Cotrufo et al. 2005b), as did litter from *Betula papyrifera* and *Populus tremuloides* from young (sapling) forest stands grown under elevated CO₂ at Aspen FACE (Parsons et al. 2004, 2008). In contrast, CO₂ growth environment did not alter subsequent litter decomposition rates for *Quercus myrtifolia* in a scrub oak community (Hall et al. 2006), for five tree species at the FACTS-1 FACE site (Finzi et al. 2001), or for *B. papyrifera* and *P. tremuloides* in older, closed-canopy stands at Aspen FACE (Liu et al. 2009). Diverse responses among experiments have been attributed to a number of factors, including soil fertility, decomposition environment, CO₂ exposure mechanism, study duration, stand development, and, genotype- and species-specific effects (Norby et al. 2001; Kasurinen et al. 2006; Hall et al. 2006; Liu et al. 2009).

Relatively little research has investigated the effects of atmospheric O₃ on litter quality and decomposition. Growth of three species of deciduous tree seedlings (Boerner and Rebbeck 1995; Scherzer et al. 1998) and two species of *Pinus* seedlings and saplings (Scherzer et al. 1998; Kainulainen et al. 2003) under elevated O₃ did not influence subsequent decomposition rates of leaf litter. Findlay et al. (1996), however, found that cottonwood leaves produced under high O₃ levels exhibited reduced decomposition rates (in aquatic microcosms), which were linked to increased phenolic concentrations. Similarly, Kasurinen et al. (2006) observed reduced decomposition in *Betula pendula* leaves produced under high O₃ concentrations. Parsons et al. (2008) found that early in stand development at Aspen FACE, growth under elevated O₃ reduced 2-year decomposition rates of *Populus tremuloides* litter, independent of CO₂ treatment. In contrast, elevated O₃ accelerated decomposition of *Betula papyrifera* litter from ambient CO₂ treatments, but decreased decomposition of litter from enriched CO₂ treatments (a significant O₃ × CO₂ interaction). Later work at Aspen FACE, however, showed that elevated O₃ decreased the decomposition of *P. tremuloides* and *B. papyrifera* litter from closed-canopy stands during only the first year of a 2.6-year study (Liu et al. 2009). As is the case for atmospheric CO₂, the effects of elevated O₃ on litter decomposition appear to be species-, age-, and context-specific.

Although too early for definitive conclusions, the developing scientific consensus is that atmospheric CO₂ and O₃ will affect litter decomposition and subsequent nutrient cycling more through their effects on the quantity, than quality, of foliage produced, and by shifting the species composition of forests (Finzi and Schlesinger 2002; Cotrufo et al. 2005b; Liu et al. 2007, 2009; Hillstrom, Meehan, Kelly and Lindroth, unpublished data). For example, soil carbon sequestration at Aspen FACE appears to be more closely related to CO₂- and O₃-mediated changes in input quantity and species composition than to

qualitative changes in litter chemical composition (Loya et al. 2003; Liu et al. 2007, 2009).

Conclusions and Recommendations for Future Research

Human-induced changes in the levels of atmospheric CO₂ and O₃ clearly influence the quantity and quality of primary production. In turn, changes in plant chemical composition cascade through above- and below-ground ecological interactions (e.g., herbivory, decomposition) to alter ecosystem structure and function, ultimately feeding back to affect terrestrial carbon sequestration and atmospheric composition. Many of these interactions are governed by processes that are fundamentally chemical in nature (double entendre intended). Thus, chemical ecologists have much to offer in terms of elucidating the mechanisms that integrate biological organization—from genomes to ecosystems—in a globally changing world.

An admittedly frustrating theme to emerge from this and other recent reviews (e.g., Stiling and Cornelissen 2007; Bidart-Bouzat and Imeh-Nathaniel 2008; Tylianakis et al. 2008) of global environmental change is that the effects of any single change factor on any particular ecological interaction are highly variable. Responses tend to be species- and context-specific, and thus appear idiosyncratic. Global change science must advance considerably further before predictions about impacts on natural systems can be made with accuracy and precision. The work of chemical ecologists should be central to such efforts: it will provide mechanistic insight into ecosystem function and provide data critical to the accurate parameterization of global change and risk assessment models.

Below I provide a non-exhaustive list of some of the more pressing needs for the contributions of chemical ecologists toward furthering our understanding of the effects of CO₂ and O₃ on forest ecosystems. Funding of large, long-term, multi-investigator projects is essential to the success of these efforts.

1. Increase the diversity of natural products studied. Our understanding of the effects of global change on the phytochemistry of forest ecosystems is based on a few classes of chemical compounds, primarily phenolics and terpenoids. Almost nothing is known about effects on other classes of secondary metabolites important to at least some tree species, e.g., alkaloids, cyanogenic glycosides, nonprotein amino acids, coumarins, and iridoid glycosides.
2. Broaden the representation of species and biomes investigated. To date, research has focused on a narrow range of deciduous and coniferous tree species and biomes, and mostly lepidopteran herbivores. Both

tropical and boreal forests are critically important to the global carbon cycle, and poorly represented in global change research. Future studies should emphasize foundation tree species (Ellison et al. 2005) and functional groups (e.g., insect borers, miners) and taxa (e.g., mammals) known to be important to the health and sustainability of forest ecosystems. The coniferous forests and irruptive bark beetles of western North America are a prime example (Raffa et al. 2008). In a similar vein, future research should address non-exploitative ecological interactions, such as mutualistic associations of plants with pollinators and mycorrhizae.

3. Emphasize simultaneous investigation of multiple global change drivers. As documented in this review and others (e.g., Tylianakis et al. 2008), the effects of any particular global change factor on ecological interactions are context-dependent. Multiple drivers should be investigated simultaneously to more realistically approximate future environments and to provide a diversity of conditions under which to test the effects of individual drivers (Tylianakis et al. 2008). Altered temperature and precipitation are arguably the most important additional factors to evaluate in the context of CO₂ and O₃ effects on forests.
4. Conduct long-term experiments. The majority of studies published to date have been executed over short time frames (up to several years), with only a few approaching 10 years—considerably less than the lifespan of forest trees. Some ecosystem processes (e.g., soil nutrient depletion and carbon storage) require assessment over the course of a decade or longer, logistical and funding constraints notwithstanding. Long-term experiments also could allow for studies of evolutionary responses of insects to CO₂- and O₃-mediated changes in tree chemistry, which are conspicuously absent in the published literature and important to understanding the effects of global change on forest ecosystems.
5. Employ “genes to ecosystems” research perspectives. “Genes to ecosystems” research has emerged as a new disciplinary frontier in genetics and ecology (Whitham et al. 2006). It combines genetic/genomic tools with the approaches of evolutionary ecology to understand the functional significance of genes in ecological context and the dynamics of ecosystems in evolutionary context. The discipline of chemical ecology can play a key role in elucidating the “metabolomes” that link gene expression to ecosystem processes in the context of global environmental change.

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