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Air quality in natural areas: Interface between the public, science and regulation

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Appropriate endpoints and exposure–response indices can improve assessment of air pollutant risk to forests in natural areas.

Abstract

Natural areas are important interfaces between air quality, the public, science and regulation. In the United States and Canada, national parks received over 315 million visits during 2004. Many natural areas have been experiencing decreased visibility, increased ozone (O_3) levels and elevated nitrogen deposition. Ozone is the most pervasive air pollutant in North American natural areas. There is an extensive scientific literature on O_3 exposure–tree response in chambered environments and, lately, free-air exposure systems. Yet, less is known about O_3 impacts on natural terrestrial ecosystems. To advance scientifically defensible O_3 risk assessment for natural forest areas, species-level measurement endpoints must be socially, economically and ecologically relevant. Exposure-based indices, based on appropriate final endpoints, present an underused opportunity to meet this need. Exposure–plant indices should have a high degree of statistical significance, have high goodness of fit, be biologically plausible and include confidence intervals to define uncertainty. They must be supported by exposure–response functions and be easy to use within an air quality regulation context. Ozone exposure–response indices developed within an ambient air context have great potential for improving risk assessment in natural forest areas and enhancing scientific literacy.

Keywords: Natural areas; Air pollutants; Ozone; Forests; Risk assessment

1. Introduction

1.1. Background

Webster's Third New International Dictionary defines a natural area as a "geographic area … having a physical and cultural individuality developed through natural growth rather than design or planning" (Gove, 1993). To the public, natural areas are embodied in national parks, national forests, protected areas, wilderness areas, ecological reserves and wildlife refuges. Yellowstone National Park, the world's first national park, was created in 1872 in the United States (US). Banff National Park, the world's third, was established in 1883 in Canada. Since their establishment over 100 years ago, the pace of demographic and economic growth in North America has been rapid, especially since the 1950s. One of the consequences of this surge in growth has been increasing background levels of air pollution (Shriner and Karnosky, 2003).

In Canada, forests cover 45% (417 M ha) of the country. Canada is in the unique position of having most of its forests under public ownership (77% provincial, 16% federal). Only 6% of Canada's forests are privately owned (Natural Resources Canada, 2006). The National Parks System in Canada is made up of National Parks and National Park Reserves, comprising 2.5% of the nation's lands and freshwater. In Canada "National parks protect natural environments representative of Canada's

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natural heritage and are protected for public understanding, appreciation and enjoyment, while being maintained in an unimpaired state for future generations'' (Parks Canada, 2006). In 2004, 40 Canadian national parks received 16 M visits at a time when the Canadian population was approximately 32 M people.

In the United States, national parks have been called ''America's greatest university without walls'' (NPS, 2006b). Preserving national parks unimpaired for the enjoyment of future generations is the fundamental purpose of the National Park Service (NPS). There are 142 national forests covering 78 M ha. In 2004, there were 300 M visits to US national parks. Under the Clean Air Act, many ''natural areas'' are designated as Class 1 areas having special national or regional natural, scenic, recreational, or historic value for which the Prevention of Significant Deterioration (PSD) regulations provide special protection. In 1997, Class I area designations were given to 158 areas that included: (1) national parks greater than 24.3 km^2; (2) national wilderness areas and national memorial parks greater than 20.2 km^2; and (3) one international park, Roosevelt–Campobello at the Canadian–US border.

1.2. Prominent air quality issues

Various air pollutants can cause damage to sensitive biological resources in natural areas. In Canadian national parks, serious threats emanate from stresses originating both inside and outside the parks, including habitat loss and fragmentation, loss of large carnivores, air pollution, pesticides, exotic species and overuse (Parks Canada, 2004). In US national parks, air pollution is considered to affect many resources and values.

There are four main air quality issues of concern to natural areas: visibility, ozone (O_3), wet deposition (including mercury (Hg)) and dry deposition of air pollutants. In the US, these issues are monitored through various networks and programs. In 1985, a national visibility monitoring program was established, the Interagency Monitoring of Protected Visual Environments (IMPROVE). Mass and chemical composition of suspended fine particulate matter ($PM_{2.5}$) and the mass of coarse particulate matter (PM_{10}) at 50 units are currently measured. Ground-level O_3 (and meteorology) is monitored by the Clean Air Status and Trends Network (CASTNet) at 34 active air-quality monitoring stations in 28 different national parks, in addition to 13 additional air-quality monitoring stations operated by states or other agencies. Trends in visibility and O_3 are discussed below.

Other air pollutants impinging on natural areas include sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and heavy metals such as Hg. Wet deposition of cations (e.g., NH_3, calcium (Ca), magnesium (Mg)), anions (e.g., sulfates (SO_4), nitrates (NO_3)) and Hg occurs when pollutants are deposited in precipitation (rain and snow) at higher elevations in clouds, and along coastal areas in fog. Wet deposition is measured through the National Atmospheric Deposition Program (NADP). Gaseous air pollutants (e.g., SO_2, NH_3), particulate pollution and vapor forms (e.g., nitric acid (HNO_3)) are deposited by complex processes such as settling, impaction and adsorption. Dry deposition in US national parks is monitored through CASTNet. Weekly average air concentrations of SO_4, NO_3, NH_3, SO_2 and HNO_3, and hourly O_3 concentrations are measured at 70 monitoring stations including 30 operated by the NPS.

Along with O_3, wet- and dry-deposited N is a major concern for forest ecosystem function and health in natural areas. In the southwest, dry deposition is the primary mode of N transfer to forests. In the northeast, wet N (and S) deposition is a regional issue. Anthropogenic sources of fixed N are having unintended consequences in terrestrial and other ecosystems. One of the consequences of excess N deposition to certain forest ecosystems can be N saturation, or the long-term removal of N limitation on biotic activity accompanied by a decrease in N retention capacity (Fenn et al., 2003). In the western US, elevated levels of N deposition increase drought stress of trees, cause buildup of dead biomass and contribute to stream and surface water contamination with NO_3 (Fenn et al., 2003).

Most air pollution that impacts park resources is emitted from sources outside of the parks. However, air pollution is also emitted as a result of various in-park activities (e.g., visitor automobiles, wildfires) (NPS, 2007). Emission inventories for 21 NPS units during 2000–2001 showed that within-unit emissions from stationary sources were, in order of decreasing amount (kg $year^{-1}$): carbon dioxide (CO_2) 28,008,573; nitrogen oxides (NO_x) 73,423; sulfur dioxide (SO_2) 52,277; volatile organic compounds (VOC) 41,563; carbon monoxide(CO) 16,895; and mass of coarse particulate matter (PM_{10}) 4290 (NPS, 2003).

1.3. Objective

In this paper, we present the concept that air quality in natural areas is an important interface between the public, science and regulation. To do this in the space available, we focus exclusively on the most pervasive air pollutant, ground-level ozone (O_3), in national parks and forests. We: (1) review the historical basis for concern; (2) present criteria for appropriate selection of measurement endpoints; (3) discuss O_3 exposure-based indices as tools for risk analysis; and, (4) advance the case for enhanced scientific literacy leading to improved risk assessment and support for essential science.

2. Air quality in natural areas

2.1. Trends in demographics

Despite having large land areas, Canada (79.4%) and the US (79%) have become highly urbanized nations (Statistics Canada, 2001; Hobbs and Stoops, 2002). The recent trend toward increased urbanization is likely to continue. The percentage of the coterminous US classified as urban increased from 2.5% in 1990 to 3.1% in 2000, an area about the size of Vermont and New Hampshire combined. (Nowak et al., 2005). Coincidentally, atmospheric transport of air pollutants and O_3 precursors away from expanding urban areas in the US and Canada to natural areas is now a reality that air quality and natural resource managers must contend with. In their thoughtful review on O_3 and natural systems, Laurence and Andersen (2003) concluded that assessing O_3 effects continues to be one of the greatest challenges in exposure–response research. The rate of national park visitation by the Canadian (~0.5 per capita in 2004) and US (~0.93 per capita in 2004) public is quite extraordinary. This means that natural areas have become ''lightning rods of public opinion and concern, complicating assessment activities even more'' (Laurence and Andersen, 2003).

2.2. Trends in two air-quality issues

2.2.1. Visibility

National parks are ''natural areas'' where forests, the public and changing air quality merge. Visibility, ecosystem health and human health issues come together to raise the level of concern and lead to political pressure and regulatory action (emissions reductions). The ability to appreciate scenic vistas greatly depends on good visibility. Particulate matter pollution is the major cause of reduced visibility in parts of the US, including many national parks (Malm, 1999). The US government, through IMPROVE, has been monitoring visibility in national parks and wilderness areas since 1988. In US scenic areas, the visual range has been substantially reduced by air pollution. In eastern parks, average visual range has decreased from 145 km to 24–40 km; one-third the visual range under natural conditions. In the west, visual range has decreased from 225 km to 56–145 km; about one-half of what it would be under natural conditions (US Environmental Pollution Agency (US EPA, 2006a). Poor visibility caused by air pollution may also point to other impacts on resources that cannot be as easily observed.

2.2.2. Ozone

Ground-level O_3 is a secondary air pollutant resulting from chemical reactions between precursor oxides of nitrogen (NO_x) and VOCs in the presence of sunlight. Variation in weather conditions plays an important role in determining ambient O_3 levels. Ozone is more readily formed on warm, sunny days when the air is stagnant. Unfortunately, such meteorological conditions dominate during the late spring–summer period (''ozone season'') when most public visits to national parks occur. At the global terrestrial biosphere–troposphere interface, O_3 is the most pervasive air pollutant. Three trends in O_3 levels have been reported by Percy et al. (2003): (1) increasing worldwide extent of potentially damaging concentrations; (2) decrease in frequency of higher peak average hourly concentrations; and (3) increasing ''background'' levels. The third trend is highly relevant to the theme of air quality in natural areas. Historical O_3 levels across North America are estimated to have offset US carbon sequestration by 18–38 Tg C year^{-1} since the 1950s (Felzer et al., 2004).

Using the nationwide network of O_3 monitoring sites, the US EPA has shown that, since the 1980s, average O_3 levels have decreased (US EPA, 2006b). However, the rate of decrease for 8 h levels slowed or leveled off during the 1990s (Fig. 1). There is new evidence that O_3 concentrations at mid-latitude sites in North America may have peaked in recent years (Oltmans et al., 2006). The NPS has analyzed air-quality trends covering the period 1995–2004. Fig. 2 presents results of the trend analysis for all indicators and reporting parks (NPS, 2006a). For O_3 (see key, Fig. 2), 16 of 32 national parks reporting had statistically significant trends (12 degrading, 4 improving). Fourteen national parks reporting showed no trend.

2.2.3. Other air pollutants

Other co-measured air pollutants in US national parks include SO_4, NO_3-N and NH_3-N. For SO_4 in precipitation (see key, Fig. 2), six of 29 national parks reporting had statistically significant trends (four improving, two degrading) (Fig. 2). Eighteen parks showed no trend in SO_4. For NO_3-N in precipitation (see key, Fig. 2), six of 29 national parks had statistically significant trends (five degrading, one improving). Sixteen parks showed no trend in NO_3-N. For NH_3-N in precipitation (see key, Fig. 2), seven of 29 national parks had statistically significant trends (all degrading). Twelve parks showed no trend in NH_3-N (NPS, 2006a).

3. Ozone and forest ecosystems

3.1. Historical basis for O_3 damage to forest trees in natural areas

Karnosky et al. (2007) have provided a 50-year retrospective view of O_3 and forests in the US. The four ''key historical events'' identified in their review had their origins in whole, or in part, in research conducted in national parks and national forests. Event 1 was the discovery of O_3's phytotoxicity to forest trees. Some of the earliest investigations on the cause of ''white pine blight'' in the southeast, including the George Washington National Forest, were conducted by Toole (1949). In the southwest Miller et al. (1963) showed that the mysterious ''X'' disease of ponderosa pine (*Pinus ponderosa* Laws.) observed in the San Bernardino National Forest was caused by photochemical oxidants (primarily O_3). Later in the east, chlorotic dwarf blight and emergence tip burn of eastern white pine (*Pinus strobus* L.) were linked to O_3. In the 1970s, Hayes and Skelly (1977) correlated increased O_3 with white pine foliar injury in the Shenandoah National Park.

Event 2 was the demonstration of population changes related to O_3 in North American forest trees. Research by Berrang et al. (1986) on wild populations of trembling aspen (*Populus tremuloides* Michx.) in national parks and recreation areas in five states (Minnesota, Michigan, Ohio, New York, Maine) documented that trees growing in areas experiencing higher background O_3 levels had been ''naturally selected'' (stage 1 of natural selection) for increased O_3 tolerance in the ambient environment.

Event 3 was the demonstration that ambient O_3 decreases tree growth and productivity. Benoit et al. (1982) extended the work of Hayes and Skelly (1977) and demonstrated that mean radial increment during 1955–1978 was smaller in O_3-sensitive white pine than in more O_3-tolerant trees. Later,

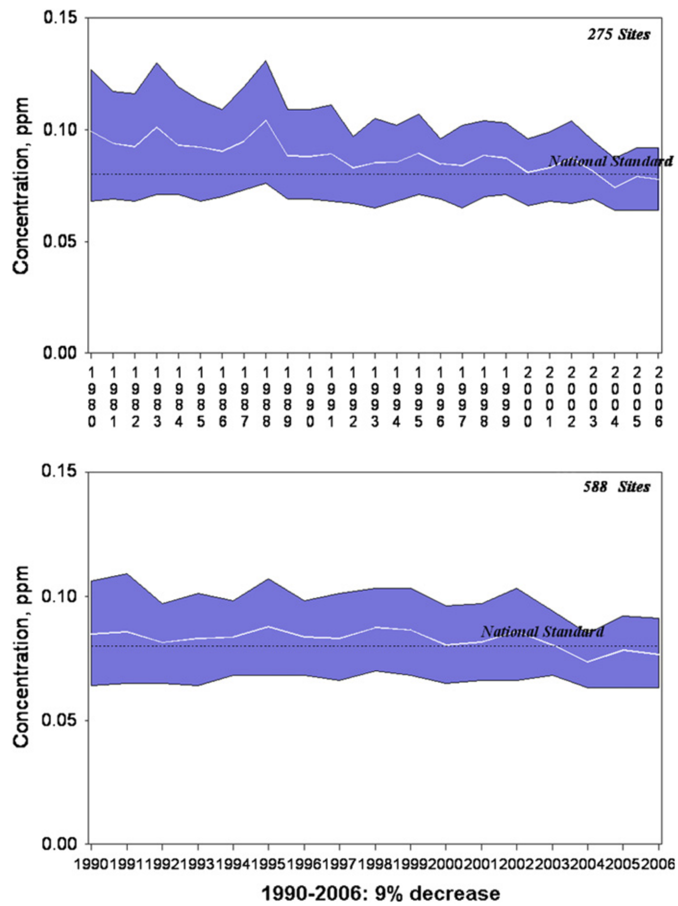

Fig. 1. National trends in O_3 air quality in the United States based on the annual 4th highest maximum 8 h O_3 concentration. The top graph shows the 8% decrease in the national average (286 sites) O_3 concentration during 1980–2005. The bottom graph shows the 8% decrease in the national average (612 sites) O_3 concentration during 1990–2005. The blue bands show the middle 80% distribution of O_3 levels among the sites. The white lines represent the average among the sites. Ninety percent of sites have concentrations below the top line, whereas 10% of sites have concentrations below the bottom line. Reproduced from US EPA (2006b).

McLaughlin and Downing (1995) used dendrometer bands on trees growing in eastern Tennessee near Great Smoky Mountains National Park to demonstrate the interactive effects of climate and O_3 on the growth of mature trees.

Event 4 is the link between O_3 exposure and community change. Replacement of the more O_3-sensitive species ponderosa pine by the more O_3-tolerant white fir (*Abies concolor* Gord. and Glend.) was first reported by Miller (1973) in the San Bernardino National Forest.

3.2. San Bernardino Mountains case study

The seminal long-term case study of the San Bernardino Mountains (SBM, including the San Bernardino National Forest) that was initiated by the late P.R. Miller in the 1960s has been comprehensively summarized. Miller and McBride (1999) brought together the collective knowledge of researchers who have worked together for up to 30 years in an effort to improve understanding of the structure and function of this mixed conifer forest ecosystem and its responses to climate and long-term exposure to photochemical oxidant air pollution. The decline of ponderosa pine first noted in the 1950s was caused by O_3 exposure. Tree mortality peaked during the drought years of the 1970s. Although air pollution levels declined during the 1980s and 1990s, foliar injury symptoms were still evident and some mortality continued.

According to McBride and Miller (1999), the dynamics, composition and structure of vegetation types in the SBM have been influenced by a changing pattern of fire frequency, intensity and extent in the 20th century, coupled more recently with oxidant air pollution and increasing levels of nitrogen (N) deposition. Long-term exposure of the SBM forests to air pollutants has demonstrated that: (1) the response of the dominant tree species (especially ponderosa and Jeffrey (*Pinus jeffreyi* Grev. and Balf.) pines) is a function of the physiological uptake of O_3 through the stomata and the impact of pollutants on leaf surfaces; (2) reduced photosynthetic productivity limits carbohydrates available for tree growth; (3) growth increments of ponderosa and Jeffrey pines as well as big cone Douglas-fir (*Pseudotsuga macrocarpa* (Vasey) Mayr.) are reduced under chronic levels of oxidant air pollution; (4) decreased crown vigor and tree productivity predisposes ponderosa pine to attack by the western bark beetle (*Dendroctonus brevicornis* LeConte); (5) the reduction of cone and seed production in

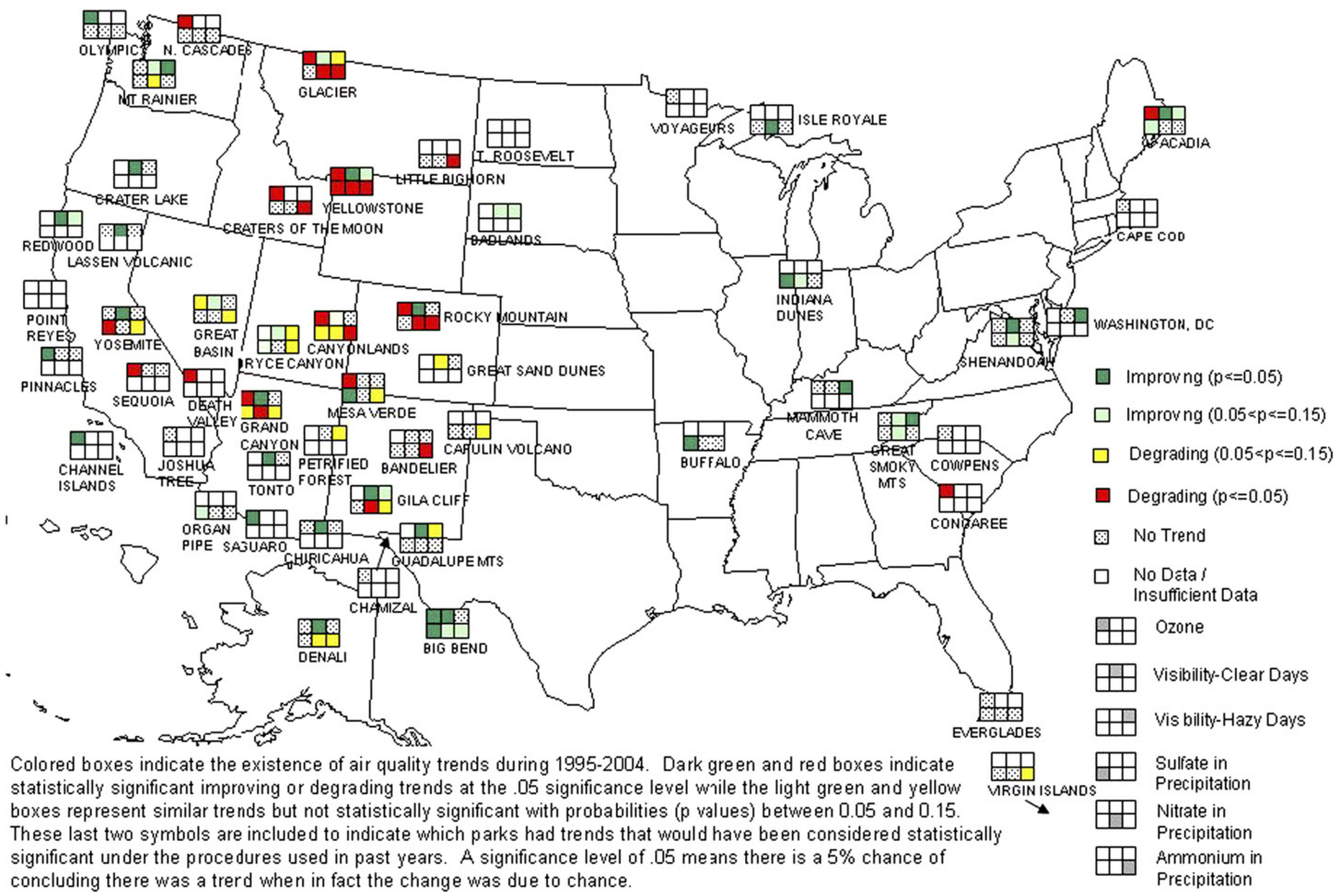

Fig. 2. Air-quality trends in US national parks, 1995–2004. The green and red boxes represent statistically significant ($p < 0.05$) improving or degrading trends during 1995–2004. The light green and yellow boxes represent similar trends that were not statistically significant at the 0.05 level. Boxes with no trends are filled with dots. Reproduced from National Park Service (2006).

ponderosa pine implies a shift in dominance toward white fir and incense cedar (*Librocedrus decurrens* Torr.) under conditions of chronic air pollution; (6) increased wet and dry N input into the system has affected the N cycle and increased N in forest soils; and (7) future interactions with global climate change (increased temperature, decreased rainfall) are to be anticipated.

The SBM long-term ecological research was subsequently extended to pattern-oriented, process investigations on O_3 distribution, N deposition and their effects in the Sierra Nevada range. This new case study includes Sequoia and Yosemite National Parks and several national forests, and is summarized in Bytnerowicz et al. (2003). Analysis of major international air pollution–forest health studies (Percy, 2002) has concluded that case studies such as the SMB have succeeded because: (1) network-level monitoring was linked to process-oriented research across spatial and temporal scales of the stressors; (2) appropriate indicators of ecosystem function were investigated at an intensity/extent appropriate to the stressors and the ecosystem; (3) investigations in essential processes/cycles were integrated with investigations into ecosystem resilience through pests, genetics, succession, etc; and (4) there was continuity in investigation. One of the elements critical to the success of both of these case studies was the coupling in space and time of air quality and climatology measurements to effects analyses (Karnosky et al., 2003a).

3.3. Ozone and natural forest plant communities

One of the under-investigated areas of O_3 impact on natural forests is the herbaceous plant community. Native herbs and shrubs are important components of forest ecosystems in natural communities. Their response to O_3 was first studied by Treshow and Stewart (1973). Since then, a number of species have become widely used as key sentinels and detectors of O_3 injury in natural areas (Krupa et al., 1998). Ambient O_3 levels in natural areas can induce visible foliar injury in sensitive herbaceous species (Souza et al., 2006), alter community composition (Barbo et al., 1998) and affect reproduction (Chappelka, 2002). As with tree species, whole-plant response to ambient O_3 in herbaceous species, such as coneflowers growing in Great Smoky Mountains National Park, is a ‘‘complex of independently varying attributes that result in varying degrees of sensitivity among individual plants’’ (Grulke et al., 2007). Within the wider context of linking O_3 impacts to nutritive quality of natural plant communities, there is new evidence that exposure to elevated levels of O_3 reduces in vitro digestibility of clover, and that co-exposure to elevated levels of CO_2 does not offset the negative effect of O_3 on nutritive quality (Muntifering et al., 2006).

3.4. Ozone–climate interaction

Assessment of forest health in North America has already made a convincing case for interaction between physical (temperature, precipitation patterns) and chemical (air pollution) climates. McLaughlin and Percy (1999) concluded that: (1) air pollutants had caused changes to C fixation and allocation to roots and shoots, and alterations in nutrient supply and changes in sensitivity to water stress had been demonstrated for several intensively studied tree species from diverse forest ecosystems across a wide geographic area; and (2) such changes had implications for how these forests could be expected to respond to climatic stresses. New research by McLauglin et al. (2007a) in eastern Tennessee near Great Smoky Mountains National Park has shown that ambient O_3 episodically increased the rate of water use and limited growth of mature trees within the study region. Stem growth loss was as great as 30–50% in a high-O_3 year, and O_3 was suggested to amplify the adverse effects of increasing temperatures on forest growth and forest hydrology (McLaughlin et al., 2007a). Indeed, McLaughlin et al. (2007b) demonstrate that ambient O_3 levels caused increases in whole-tree canopy conductance and depletion of soil moisture in the rooting zone, and reduced late-season streamflow in forested watersheds. This insightful research is perhaps the first to make a definitive link between current ambient O_3 exposures (air quality) and the frequency and level of negative effects of drought (climate) on forest hydrology.

4. Research methods, endpoints and exposure indices

4.1. Research methods used

Research methods used to investigate O_3 exposure–tree response relationships have increased in scale, paralleling the key historical events (Karnosky et al., 2007). Until the mid-1990s, O_3 exposure–response science was conducted in laboratory laminar flow cabinets, growth chambers, continuous-stirred reactor (CSTR) chambers, and open-top chambers (OTC). These chambered studies were constrained by size and growing space and necessitated the use of seedling or young sapling stage plant material. With the advent of free-air exposure techniques in the mid 1990s (Hendrey et al., 1999), it became feasible to study cellular to stand-level effects at different trophic levels within the ambient air context. Seven years (1998–2004) of free-air O_3 exposure with northern hardwood species at the Aspen Free Air Carbon Dioxide Enrichment (FACE) site in Rhinelander, Wisconsin have shown that: (1) O_3 effects cascade through tree gene expression, biochemistry and physiology; (2) these primary effects feed back to productivity; (3) "bottom-up" changes to foliar biochemistry and physiology predispose the trees to pest attack; and (4) tree-level changes result in changes to element (C, N) cycling and water-use efficiency (Percy et al., 2002; Karnosky et al., 2003c, 2005; King et. al., 2005).

The Aspen FACE experiment has confirmed earlier OTC findings on productivity loss under O_3. However, it has not as yet (biomass harvests following 3 and 5 years exposure to O_3) provided confirmation for previously reported enclosed chamber (OTC) changes in allometry and carbon allocation (King et al., 2005 and Kubiske et al., 2006). It is quite possible that OTC studies may have overestimated the negative O_3 impact on allometry and carbon allocation. Long et al. (2006) have recently demonstrated that (at least in the case of elevated CO_2 in FACE experiments) soybean crop yield enhancement in free-air rings was ~50% less than in enclosed chamber studies. In fact, for three key production measures (yield, biomass, photosynthesis) in four crops, only one of the 12 items was not lower in FACE exposure than in the chamber equivalent.

Uniquely, one team of investigators (Karnosky et al., 2006) has used identical sets of trembling aspen genetic material (clone 216, intermediate in O_3 tolerance; clone 259, O_3 sensitive; and clone 271, O_3 tolerant) in three growth and productivity studies. The studies were done in OTC, a free-air system (Aspen FACE), and field plots growing under differing levels of ambient O_3 in the US Lake States. The results showed that the direction of response was remarkably consistent across the OTC, free-air and ambient plot studies (Table 1). Ozone induced visible foliar symptoms (black bifacial stipple or larger necrotic areas), decreased leaf retention, altered epicuticular wax chemistry and structure, decreased photosynthesis and increased stomatal conductance in the O_3-sensitive

Table 1
Significant responses shown for trembling aspen exposed to elevated O_3 in open-top chambers (OTC), a free-air exposure system (FACE) and an ambient O_3 plot in the Great Lakes States Region (Ambient O_3) (modified from Karnosky et al., 2006a)

Measurement	OTC	FACE	Ambient O_3
Leaf condition			
Visible symptoms	*	*	*
Premature leaf abscission	*	*	*
Epicuticular waxes	–	*	*
Pest occurrence	–	*	*
Gene expression	*	*	–
Photosynthesis	*	*	*
Stomatal conductance	*	*	*
Growth and productivity			
Height and diameter	*	*	*
Biomass	*	*	–
Root growth	*	*	–
Stand dynamics	–	*	*
Essential cycles			
Carbon	–	*	–
Nitrogen	–	*	–
Water	–	*	–
Fitness			
Clonal variation	*	*	*
Survival	–	*	*
Reproduction	–	*	*

*, statistically significant O_3 effect found; –, not evaluated.

genotypes in all three sets of studies. The clonal rankings of O_3 sensitivity were maintained across the OTC (Karnosky et al., 1996), Aspen FACE (Karnosky et al., 2003c) and ambient O_3 plot studies (Karnosky et al., 2003b). Averaged across the clones, O_3 exposure decreased productivity between 29.3% (OTC study, Karnosky et al., 1996) and 23% (Aspen FACE, King et al., 2005).

4.2. Extrapolation of O_3 exposure–response research

There is an extensive literature on O_3 exposure–tree response in chambered environments (Karnosky et al., 2007). Although great progress has been made in understanding O_3 effects on agricultural crops and individual forest species, much less is known about impacts on natural terrestrial ecosystems (Laurence and Andersen, 2003). Although critical to advancing our understanding on mechanisms of action, a limitation of the literature from chambered studies is the inability of air-quality regulators to extrapolate much of this very good science to risk assessment. This is best evidenced in the 6–7 December 2005 Clean Air Scientific Advisory Committee (CASAC) Peer Review of the US EPA Air Quality Criteria for Ozone and Related Photochemical Oxidants (2nd External Review Draft). Quoting from review comments published on Section AX9-5-4-5, Scaling Experimental Data to Field Conditions (natural areas), CASAC stated that "This does not mean to suggest that OTC results are not good science but rather that the extrapolation of the OTC results to 'quantitatively' reflect the response of the vegetation in the ambient environment is questionable" (CASAC, 2006).

In his review, Manning (2005a) had previously stated that much of the data derived from chambered environments appears to have limited use for extrapolation to risk analysis because of differences in climatic and growth environment between chambered and ambient air situations. Chambered OTC environments have been shown to affect plant growth and development, which, together, are influential in determining plant response to ambient O_3 (Elagöz and Manning, 2005). Modified temperatures and air movement within OTCs (filtered and non-filtered) are known to increase growth of apple seedlings and change the incidence of disease and arthropod pests compared with trees grown in ambient air treatments (Manning et al., 2004). There are also a number of key ecosystem processes that cannot be studied in chambered O_3 exposure systems (Table 1). Insect and disease incidence and severity, stand dynamics, element (C, N) cycling, water-use efficiency and reproduction all contribute to overall ecosystem health and function, and are important co-factors in tree response to O_3. So, the question remains: were the research methods and endpoints appropriate for risk analysis within an ambient air context?

4.3. Measurement endpoints

An endpoint is defined as "the point marking the completion of a process or stage of a process: *the final point*" (Gove, 1993).What endpoints are appropriate for scientifically defensible O_3 exposure–response estimation in natural areas? In the case of visibility, degree of light scattering by air pollutants (mainly fine particulate matter) is an appropriate endpoint. It is the *final point* of many complex biogenic and anthropogenic atmospheric physicochemical interactions. It is quantifiable and routinely monitored in many national parks. Visual images are widely available through the multi-agency cooperative webcam facilities (see http://www.epa.gov/airtrends). Trends in degree and pattern of change are widely distributed and easily understood by the public. Decreased visibility reduces aesthetic value; improved visibility enhances it. Public enjoyment of natural areas and cost:benefit for air-quality management can be quantitatively associated with degree of change in the final endpoint. For instance, the Clean Air Act Amendments of 1990 are predicted to exceed their costs by a margin of four to one. In 2010, the human health and ecological benefits of Clean Air Act programs are estimated to total about \$110 billion (US EPA, 1999).

For forests in natural areas, a chosen species-level endpoint must have social, economic and ecological relevance (Hogsett et al., 1997). However, the interaction of O_3 with trees is a nonlinear (Percy et al., 2007), and very complex process that varies in response to a host of factors including meteorology (Kubiske et al., 2006), bottom-up and top-down pest cycles (Percy et al., 2002) and stand dynamics (McDonald et al., 2002) to name a few. This presents a large challenge when scaling impacts beyond the tree level (Samuelson and Kelly, 2001).

Endpoints that have been used to assess O_3 risk to forest trees in natural areas include presence and degree of visible foliar injury, tree productivity, or change in other ecosystem attributes such as lichen density and species composition. However, there is often no commonality among responses in these endpoints when assessed coincidentally in high and low deposition areas (Muir and McCune, 1988). In this paper, we build on the work of Hogsett et al. (1997) to advance discussion on selection of endpoints appropriate for scientifically defensible O_3 exposure–response estimation in natural areas. We list below six questions relating to endpoint selection. We suggest that they be addressed by researchers, land managers, policy specialists and air-quality regulators when undertaking any endpoint selection process.

1. Is the endpoint a *final point* in a key ecosystem process?
2. Can the endpoint be measured with accuracy and, importantly, with precision over time?
3. Is the endpoint supported by published exposure–response science completed within the ambient air context?
4. If the endpoint's status changes, does this provide feedback to tree productivity or some other ecosystem value of social, economic or ecological importance?
5. If positive or negative feedback is demonstrated, is the estimate of change in a form that can be understood and used by air-quality regulators?
6. If it can be used by regulators, can it be used in the short term to support decisions on air-quality management (prevention), or in the longer term to provide scientific input to a criterion-setting process (protection)?

5. Linking air quality with endpoints in natural areas

5.1. Ozone exposure–response indices

Establishing cause–effect relationships for ambient O_3 exposure and tree growth has proved to be an elusive goal (Manning, 2005a). For human health effects, exposure-based metrics (indices) provide a valuable tool for assessing relative effectiveness of alternative control strategies (Foley et al., 2003). Exposure-based metrics also introduce a higher degree of accountability in meeting National Ambient Air Quality Standards (NAAQS) by augmenting air-quality metrics with ones more closely associated with morbidity and mortality (endpoint question #4) caused by air pollution exposure.

A considerable scientific literature on O_3 exposure–vegetation response indices exists dating back to at least Runeckles (1974). Musselman et al. (2006) have provided a thorough review of the use of hourly averaged O_3 data with plant response. Ozone exposure indices most commonly used in past analyses of exposure–response relationships were: (1) the SUM06 threshold-based sum of daytime O_3 concentrations $\geq$0.06 ppm (Lefohn and Foley, 1992); (2) the W126 sigmoidally weighted W126 function (Lefohn and Runeckles, 1987); and (3) the accumulated over a threshold (AOT) based sum of hours of the day (global radiation $>$50 W m^{-2}) with O_3 concentrations $>$40 ppb (Fuhrer et al., 1997). Recently, McLaughlin and Nosal (in press) have used a dendroecological free-air approach to model effects of O_3 in the presence of co-varying influences of other environmental variables important to O_3 flux. Model predictions of growth loss in mature southern hardwood species were in the range of 50% in high O_3 years and agreed well with observed growth. Such an approach has great potential for determining contribution of O_3 to changes measured in tree growth, and for scaling hourly effects of O_3 to cumulative impacts over the growing season (McLaughlin et al., 2003).

5.2. Current O_3 air-quality standards

In Europe, critical levels are set to protect vegetation. The current level used in O_3 risk assessment is the exposure-based AOT40. However, a flux-based concept is now being advanced for European forest trees (Matyssek et al., 2007). In North America, ambient air-quality standards are promulgated that do not imply a threshold (critical level) concentration. The current US and Canada O_3 air-quality standard is "the 3 year average of the annual fourth-highest daily maximum 8 hour average O_3 concentration" (Federal Register, 1997 and Canadian Council of Ministers of the Environment (CCME), 2000). The US has established the O_3 primary (human health-based) and secondary (welfare-based) NAAQS to be the same, with the level set at 80 ppb O_3. In Canada, the metric form and averaging times are the same as in the US, but the level differs. In Canada, the Canada Wide Standard (CWS) for Particulate Matter and Ozone (CCME, 2000) has set a human health-based level of 65 ppb O_3.

Key ongoing issues in the continuing evolution of North American O_3 air-quality standards pertain to the relative importance of higher vs. mid-range O_3 concentrations, as well as the need for a separate secondary NAAQS to protect vegetation. As part of the ongoing science review of O_3 criteria, the US EPA staff paper proposed invoking a separate secondary standard to protect vegetation from O_3 damage. They recommended a move from the current NAAQS concentration-based form to a 12 h (08:00–20:00) W126 O_3 exposure index accumulated over a 3-month period. Staff recommended a range of levels from 21 down to 7 ppm/h (US EPA, 2007). Ozone exposure–response models developed from the Aspen FACE experiment are now available (Percy et al., 2007). These models quantify uncertainty (95% confidence bands), a requirement to produce useful scientific advice to those in the policy arena (Laurence and Andersen, 2003). The regression-based models developed for trembling aspen (five clones covering a wide range of sensitivity to O_3) and white birch (*Betula papyrifera* Marsh.) comprise annual growing season 4th highest daily maximum 8 h average O_3 concentration, cumulative growing degree days (GDDs) and average windspeed. The models predict extremely well within a wide range of 4th highest daily maximum 8 h average O_3 concentrations and, thus, have immediate relevancy to ambient exposure conditions experienced by two of North America's most widely distributed (26+ M ha) tree species (Percy et al., 2006b; Percy et al., 2007).

Regression analysis using the same 5-year O_3 exposure–response data has demonstrated that the growing season 4th highest daily maximum 8 h average O_3 concentration performed much better as a single indicator of aspen cross-sectional area growth than did SUM60, AOT40, W126, or maximum 1 h average O_3 concentration (Percy et al., in press). The W126 O_3 exposure index was determined to overestimate the negative effect of O_3 on trembling aspen and white birch final endpoint (growth) response. Therefore, we believe that the growing season 4th highest daily maximum 8 h average O_3 concentration, cumulative GDDs and average windspeed-based models may provide a new opportunity for scientifically defensible risk analysis within the North American air-quality context. The models require only a change in averaging time (3 years to annual) and slight change in form from the current primary NAAQS and CWS for O_3 (Percy et al., in press), In reality, the current primary NAAQS (Federal Register, 1997) only requires data from the 2nd and 3rd quarters of the year (the "ozone season"). Ultimately, ground proofing to test biological significance of these new models (Manning, 2003) must be completed.

6. The way forward

6.1. Research

Fifty years of research has aimed at understanding plant response to O_3. Laurence and Andersen (2003) have stated that, "Many natural systems are not amenable to experimental research because of their stature, their location, or their sensitivity to human impact. However, we must understand the potential sensitivity of the systems to ozone because they are intimately involved in many essential ecosystem services." In their review of O_3 and forests in the US, Karnosky et al. (2007) listed six areas where further research on O_3 and forest response is warranted. With respect to O_3 and forests in natural areas, the adequacy of O_3 standards to protect forest ecosystems and the scaling of empirical results to landscape levels are two areas of paramount importance to land managers and air-quality regulators.

Analysis of exposure–response data from tightly coupled free-air experiments, and field measurement using dendroecological techniques conducted within the ambient air context are positive developments that can contribute much to addressing these two research needs. Their positive attributes include the continual co-measurement of O_3, meteorology, flux regulators and response in mature trees over multiple years. This permits the rigorous application of regression techniques (a very powerful statistical tool) to produce predictive models developed against a backdrop of inter-annual changes in physical climate, stand dynamics, elemental/water cycles and pest populations. The use of multiple genotypes and species in a free-air setting like Aspen FACE (Karnosky et al., 2005) or in ambient-air clonal plantations (Karnosky et al., 2003b) can also yield models of genotype–environment interaction across a wide range of species sensitivity to O_3 (Percy et al., 2007). As summarized by Manning (2005a), "while it is difficult to work in natural ambient conditions, studies under these conditions are essential to develop a better understanding of how ozone and environmental factors interact to affect radial growth of trees."

6.2. Toward increased scientific literacy

According to Orbach (2005), there are "… three imperatives of economic growth, scientific literacy and intellectual excitement, that, if properly understood by our society will, benefit us all." At the same time, we as scientists investigating exposure–response in natural areas "… must make clear how we contribute to each, providing rationale for increased support for science." Should we be concerned about air pollution in our national parks? According to Lefohn (2002), "… the answer is a definite yes! However, … to make the best decisions concerning our national parks … the public, politicians, and government officials must be exposed to the best science available." Exposure-based indices present an underused opportunity to meet these needs within the North American air-quality context. However, indices must be statistically strong, have a high goodness of fit, and include confidence intervals for us to define uncertainty in the prediction. They must be supported by exposure–response science and be based on quantifiable, biologically relevant endpoints that have social, economic and ecological relevance. They must also be simple to use within an air-quality regulatory context. Ultimately, however, if ground proofing is not done, then the standards (exposure indices) will have no biological significance and will only be exercises in air-quality assessment (Manning, 2003).

Exposure–response scientists must work more closely with the regulatory community to agree on a set of measurement endpoints that will provide the basis for accurate and defensible

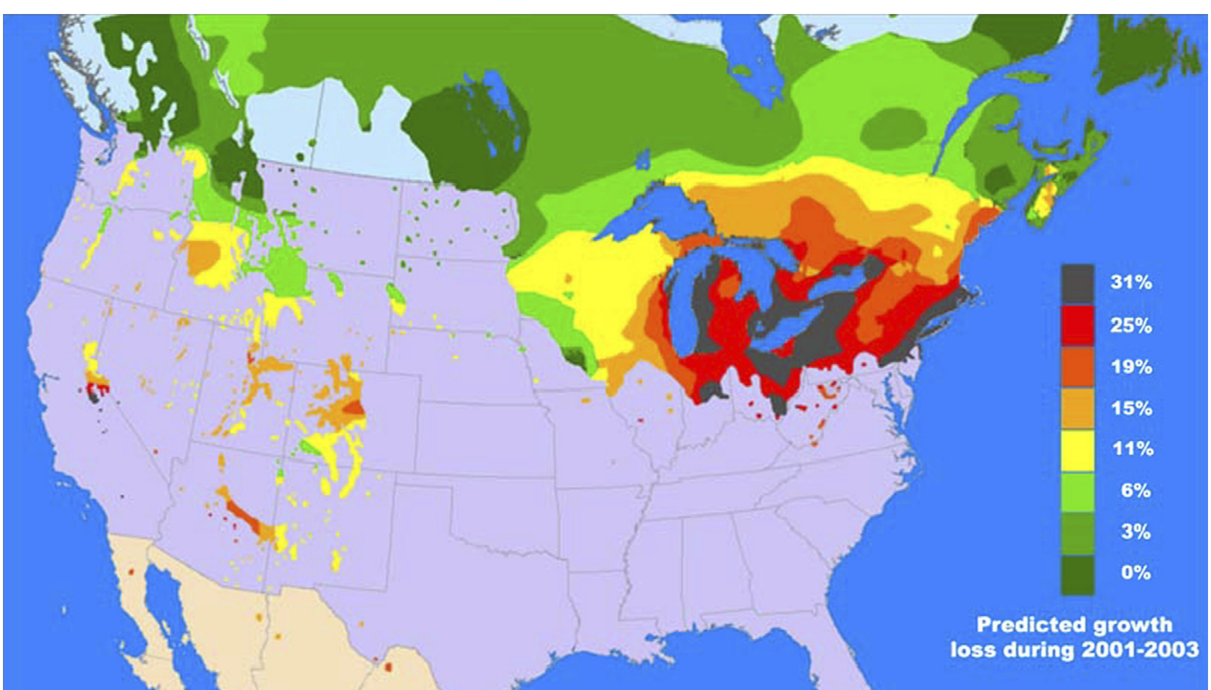

Fig. 3. Modeled trembling aspen growth loss in natural populations across North American caused by a 3-year (2001–2003) average of the ambient annual 4th highest daily maximum 8 h average O_3 concentration. Change in growth was calculated using the new exposure–response model developed from the Aspen FACE multifactor global change experiment. Areas where trembling aspen does not occur are shown in light blue (Canada), gray (USA) and light brown (Mexico). The northerly portions of the aspen range in Canada and Alaska were removed because O_3 monitoring was unavailable. Reproduced from Percy et al. (2006c).

risk assessments (Laurence and Andersen, 2003). Multifactor global change experiments can answer big science questions and contribute to enhancing scientific literacy (Percy et al., 2006a). Fig. 3 displays the predicted outcomes calculated for an appropriate endpoint after applying the exposure–response models developed from the Aspen FACE multifactor experiment within the North American ambient-air context. Change in growth was calculated for each of the clones represented. Degree of change in growth per clone at increasing 5 ppb increments of annual growing season 4th highest daily maximum 8 h average O_3 concentration was then averaged across the four clones (271, 216, 259, 42E) that responded negatively to O_3 and the clone (8L) that responded positively. This prediction (%) was finally applied to the trembling aspen spatial distribution.

Significantly, this data set (Percy et al., 2006b, 2007) conforms in principle to the concept of hormesis (Calabrese, 2005). Simply put, the exposure–response curve shape (here an inverted U shape) demonstrated lower exposure stimulation followed by higher exposure inhibition. Too often in the past, O_3 exposure–response science has focused exclusively on negative effects (Manning, 2005b). This does not reflect the natural biological reality in forest impacts or adaptation to stressors, including air pollutants or climate change.

7. Conclusions

In North America, natural areas are important interfaces between air quality, the public, science and regulation. In the US and Canada, there were over 315 million visits to national parks during 2004. Forest ecosystems in many national parks, forests and wilderness areas are being exposed to elevated levels of air pollutants that are affecting important anthropocentric and ecological values, including forest health. To date, risk assessment for O_3 and North American forests in natural areas has been constrained by experimental techniques used for exposure–plant response. Analysis of new exposure–response data from tightly coupled free-air experiments, and field measurement using dendroecological techniques conducted within the ambient-air context are now available. Exposure–response models using O_3-exposure indices developed around ambient-air quality standards have the potential to improve risk assessment, strengthen accountability, and at the same time, enhance scientific literacy, leading to increased support for science.

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