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New exposure-based metric approach for evaluating O_3 risk to North American aspen forests

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A new exposure-based metric approach to predict O_3 risk to North American aspen forests has been developed.

Abstract

The United States and Canada currently use exposure-based metrics to protect vegetation from O_3. Using 5 years (1999–2003) of co-measured O_3, meteorology and growth response, we have developed exposure-based regression models that predict *Populus tremuloides* growth change within the North American ambient air quality context. The models comprised growing season fourth-highest daily maximum 8-h average O_3 concentration, growing degree days, and wind speed. They had high statistical significance, high goodness of fit, include 95% confidence intervals for tree growth change, and are simple to use. Averaged across a wide range of clonal sensitivity, historical 2001–2003 growth change over most of the 26 M ha *P. tremuloides* distribution was estimated to have ranged from no impact (0%) to strong negative impacts (−31%). With four aspen clones responding negatively (one responded positively) to O_3, the growing season fourth-highest daily maximum 8-h average O_3 concentration performed much better than growing season SUM06, AOT40 or maximum 1 h average O_3 concentration metrics as a single indicator of aspen stem cross-sectional area growth.

Keywords: Ozone exposure-based metric; Risk prediction; Air quality standards; Trembling aspen; Growth

1. Introduction

It is widely perceived that climate change such as global warming may lead to increased growth and range distribution of some forests (Cox et al., 2000). This warming is largely being driven by increased radiative forcing caused by rising levels of greenhouse gases. The third most important greenhouse gas contributing to global average radiative forcing is tropospheric ozone (O_3) (Rawaswamy et al., 2001). In the lower troposphere, surface level O_3 has become one of the most pervasive air pollutants at the terrestrial biosphere–troposphere interface (Fowler et al., 1999). Current annual average background O_3 concentrations over mid-latitudes of the northern hemisphere range between 20 and 45 ppb and the annual cycle is characterized by a spring maximum that peaks during May (Vingarzan, 2004). In their review of variability of background O_3 in the lower troposphere, Lefohn et al. (2001) concluded that: (1) the substantial background O_3 present in the lower troposphere of the northern hemisphere is formed from both stratospheric and photochemical tropospheric sources; (2) that at more northerly latitudes, stratospheric processes play a significant role in defining these

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background O_3 concentrations; and (3) stratospheric processes may also be important in contributing to O_3 levels in the 50–60 ppb range or higher at more southerly locations.

Scenarios for changes in spatial patterns of air quality (O_3) are being driven not only by weather and climate variability, but by changes in demographics, land use, and economic growth (Shriner and Karnosky, 2003). Historical increases in anthropogenic emissions of the tropospheric ozone (O_3) precursor gases, nitrogen oxides (NO_x), and volatile organic compounds (VOCs), have led to a large increase in average, surface-level O_3 in the northern hemisphere over the past 100 years (Finlayson-Pitts and Pitts, 2000). Over mid-latitudes, background O_3 levels have risen approximately 0.5–2% per year (Vingarzan, 2004). Surface-level O_3 is a growing air pollution problem, and threatens forests in both northern and southern hemispheres (Percy et al., 2003). With increasing expansion of city centers and urban sprawl, unmanaged forest areas will increasingly become recipients of long-range transport of secondary oxidation products of primary urban VOC and NO_x emissions. Modeled estimates (Fiore et al., 2002) indicate that anthropogenic emissions in Asia and Europe may increase afternoon O_3 concentrations in surface air over the United States by between 4 and 7 ppb, an enhancement that may be particularly large for O_3 concentrations in the mid range (50–70 ppb). Any rise in background O_3 concentration may, therefore, offset the positive benefits for North American forests accrued from the recent downward trend in peak O_3 concentrations. In addition, short-term variations in O_3 exposure may lead to strong cumulative growth effects over the growing season (McLaughlin and Nosal, in press). This scenario presents a new challenge to air quality regulators.

Since the late 1950s, an extensive scientific literature has been built on the O_3 impacts on forest trees (see reviews by Kickert and Krupa, 1990; Chappelka and Samuelson, 1998; McLaughlin and Percy, 1999; Percy, 2002; Percy et al., 2003; Andersen, 2003; Ashmore, 2004; Karnosky et al., in press), forest ecosystems (Miller and McBride, 1999; Bytnerowicz et al., 2003) and physiologically based modeling of North American forest response. Ollinger et al. (1997) simulated the effects of O_3 on hardwood forest types in the northeastern US and estimated growth reductions between 3% and 22%. Later, Laurence et al. (2001) linked the mechanistic TREGRO model with the ZELIG stand model, parameterized them with biological/meteorological data from three sites, and simulated 100-year growth under five O_3 exposure regimes. Change in *Pinus taeda* basal area ranged from +44% to −87% depending on O_3 exposure and precipitation, whereas basal area of *Liriodendron tulipifera* (generally considered O_3 sensitive) was not affected. Weinstein et al. (2005) used the same models to simulate growth of *Pinus ponderosa* and *Abies concolor* under increased O_3 exposures in the western San Bernardino and Sierra Nevada mountains. They predicted negative effects on *P. ponderosa* but little response in *A. concolor* due to differential sensitivities to O_3, influences of competition, and soil moisture. Interestingly, simulations by Tingey et al. (2004) were among the first to demonstrate a link between improved emission control strategies and improved tree growth. However, there remain questions as to whether process models can be accurately parameterized to predict mature tree response (Samuelson and Kelly, 2001).

Recently, physiological effects of O_3 and biogeochemical changes have been scaled (Ollinger et al., 2002; Felzer et al., 2004) to landscape productivity. These models predict that tropospheric O_3 levels in the US can largely offset increased forest productivity due to increasing atmospheric CO_2 concentrations. Although certainly indicating the direction and magnitude of potential impact on forest productivity, these models are built partially upon assumptions around linearity of response and O_3 metrics that may not perform well within the North American ambient air context (Karnosky et al., 2005; Percy et al., in press).

In North America, the best available scientific knowledge, balanced by social, economic, and political considerations, is employed to set ambient air quality standards for regulatory purposes. The United States and Canada recently established the O_3 air quality standard metric as "the 3-year average of the annual fourth-highest daily maximum 8-h average O_3 concentration" (Federal Register, 1997; CCME, 2000). In the US, there is a primary standard (human health-based) and a secondary standard (welfare-based) that can be different or the same (Percy et al., 2003). The current US EPA primary National Ambient Air Quality Standard (NAAQS) for O_3 is set at 80 ppb. At this time, the secondary standard is set to be the same as the legally binding primary standard. In Canada, the form and averaging time are the same as the US but the level differs. In Canada, the Canada-wide Standard (CWS) for ozone (CCME, 2000) established a human health-based target level of 65 ppb O_3. Significantly, the US and Canadian standards do not assume the existence of a receptor concentration threshold (Wolff, 1996). Rather, the numerical expression for the US and Canadian O_3 standards is founded upon what is considered an adequate margin of safety based on current scientific knowledge and understanding.

When examining accountability within the current US National Ambient Air Quality Standard (NAAQS) for O_3, Foley et al. (2003) stated that, for human effects, "Exposure-based metrics provide an information-rich tool in assessing relative effectiveness of alternative control strategies and introduce a higher degree of accountability in meeting NAAQS by augmenting air quality metrics with ones more closely associated with morbidity and mortality caused by air pollution exposure." In the specific case of regulating surface-level O_3 to protect vegetation, there is an extensive literature of chamber-based [i.e., continuous-stirred reactor chamber (CSTR), open-top chamber (OTC)] studies where O_3 was found to elicit strong negative responses. Although essential to understanding mechanisms of action, a limitation of this research has been the inability of regulators to extrapolate much of this very good science for application in air quality criteria setting and risk assessment. In the final analysis, modeling work to date has relied for the most part on exposure–response and mechanistic data derived from chambered environments that, in the end, have limited utility in extrapolation to risk analysis (Manning, 2005a) due largely to differences in growth environment between chambered and non-chambered situations. This point has recently been re-stated emphatically by Long et al. (2006) who demonstrated that (in the case of elevated CO_2 in free-air experiments) crop yield enhancement in the free-air rings was ~50% less than in enclosed chamber studies. Clearly, continued research is needed to define our estimate of the level of exposure that will protect vegetation (Laurence and Andersen, 2003).

It is clear from the Musselman et al. (2006) review that, during the past 30 years, hourly averaged O_3 data have been summarized in many different ways to assess risk to vegetation. Among indices receiving the most attention in analyses of exposure–response relationships in chambered studies have been the SUM06 threshold-based sum of daytime O_3 concentrations ≥60 ppb (Lefohn and Foley, 1992) and the accumulated over-a-threshold (AOT)-based sum of hours of the day with O_3 concentrations >40 ppb and clear-sky global radiation above 50 W m^{-2} (Fuhrer et al., 1997). Recently, McLaughlin and Nosal (in press) have used a field-based open-air approach with electromechanical dendrometer techniques to model specific effects of O_3 in the presence of covarying influences of other environmental variables important to O_3 flux. Regression coefficients for ambient O_3 exposure (cumulative SUM06) prediction were negative and statistically significant for *Pinus rigida, Quercus rubra, Quercus prinus,* and *Carya* sp. Model predictions of growth loss in the range of 50% in high O_3 years agreed well with observed growth. This approach also 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 impact over the growing season (McLaughlin et al., 2003). However, preliminary regression analysis of the efficacy of a modified (growing season) version of the US–Canadian O_3 air quality standard metric form by Karnosky et al. (2005) has indicated that the growing season 4th highest daily maximum average O_3 concentration was potentially a good indicator of trembling aspen (*Populus tremuloides* Michx.) diameter growth. To date, there have been no further studies completed on the efficacy of the recently established O_3 air quality standard metric form as "the 3-year average of the annual fourth-highest daily maximum 8-h average O_3 concentration".

In their review, Musselman et al. (2006) concluded that until effective dose models are developed, "…exposure-based metrics appear to be the only practical measure for use in relating ambient air quality standards [in North America] to vegetation response." Here we build upon the earlier prediction approach of Hogsett et al. (1997). By considering an important endpoint in a key natural system species as originally suggested by Laurence and Andersen (2003), we sought to develop new exposure-based response relationships from co-measured indicator-response data collected at Aspen FACE, a multi-year, ecosystem-level, free-air exposure system designed to reflect the ambient air quality reality in North America.

The objective of this study was to develop exposure–response models based upon the formulation suggested by Krupa et al. (2003) that would then be used to: (1) retrospectively estimate 2001–2003 productivity change across the range of North America's most widely (26 M ha) distributed tree species (Burns and Honkala, 1990), trembling aspen (*Populus tremuloides* Michx.); and (2) evaluate the efficacy of several commonly used O_3 metrics as indicators of aspen growth response.

2. Materials and methods

2.1. North American surface O_3 exposure

To spatially represent the short-term historical distribution of measured ambient surface O_3 concentrations across the North American landscape, 3 years (2001–2003) of hourly average data were obtained from Environment Canada's National Air Pollution Surveillance (NAPS) network database and from the US Environmental Protection Agency Air Quality System (AQS) database. The US EPA NAAQS and Canadian CWS metric form a 3-year average of the fourth-highest daily maximum 8-h average O_3 concentrations (Federal Register, 1997; CCME, 2000) that was used to calculate O_3 concentration for each of 307 monitors located away from city centers. The distribution was created using the Map Generator Version 2.0 software created for the US EPA to drive the AIRNOW mapping application. The interpolation method selected was inverse distance weighting.

2.2. The Aspen FACE experiment

The Aspen FACE experiment (32 ha) is situated on sandy loam glacial outwash soil in northern Wisconsin, near Rhinelander (45°06′ N, 89°07′ W; 490 m a.s.l.; www.aspenface.mtu.edu). The system has been used to fumigate (1998–present) aggrading trembling aspen (five genotypes), mixed white birch (*Betula papyrifera* Marsh.)/trembling aspen and mixed sugar maple (*Acer saccharum* Marsh.)/trembling aspen stands. The experiment comprises three randomized blocks containing 12, 30-m diameter FACE rings, assigned to factorial treatments of CO_2 and O_3. Design and performance characteristics of Aspen FACE have been published and are available elsewhere (Dickson et al., 2000; Karnosky et al., 2003b, 2005; Kubiske et al., 2006). Growth, O_3, and meteorology data from a 5-year period (1999–2003) were used in this study.

2.3. Ozone fumigation treatments

Ozone was generated from oxygen (99.7% pure) using a 20-lbs per day Praxair Ozone Generator. The Aspen FACE protocol for O_3 fumigation included a 07:00–19:00 h (12 h) daily exposure, 7 days a week from bud break to bud set (136–144 day growing season during this study period). Ozone was not released if leaf surfaces were wet or if daily maximum temperature was predicted to be <15 °C. In practice (Percy et al., in press), O_3 was fumigated on only 48.7–51.6% of potential growing season days as follows: 1999 (124 days, 820 h); 2000 (121 days, 800 h); 2001 (122 days, 777 h); 2002 (107 days, 787 h); 2003 (117 days, 893 h). Target elevated O_3 was 1.4× ambient air. Daytime 90% confidence intervals were $12 \leq 48 \leq 84$ ppb for elevated O_3 based on the life of the experiment including days when O_3 was not fumigated (Kubiske et al., 2006). Details on diurnal O_3 profiles and control to elevated FACE ring comparisons are available elsewhere (Karnosky et al., 2005).

2.4. Response and indicator variables

We tested five aspen clones (pure aspen plantation, eastern half of each FACE ring; total = 1723 trees in 1999) covering a wide range of documented sensitivity (Karnosky et al., 2005) to O_3. We built a matrix of 30 cases [5 years' data × 6 FACE rings (3 control, 3 O_3)] for analysis. Each case comprised: (1) a response variable (mean diameter converted to mean cross-sectional area); (2) an O_3 indicator variable (several O_3 quantifications were investigated but the best predictive model was obtained using the growing season 4th highest daily maximum 8-h average O_3 concentration); and

(3) six meteorological or flux regulator variables important in controlling ambient O_3 concentrations and O_3 uptake by plants (NRC, 1991; Krupa et al., 2003).

2.4.1. Response variable

End of growing season tree diameters were measured at 3 cm (1998–2001) or at 10 cm (2001–2003) above ground. Diameters for 2001 used were the average of 3 cm and 10 cm as described by Kubiske et al. (2006). All measurements were collected on trees growing within the core area (∼5 rows inward from the free-air inlets toward the ring center) where elevated O_3 was most stable. Diameters (dia. ± 1 cm) were converted to cross-sectional area using the commonly used equation: cross-sectional area (m^2) = 0.00007854 × ($dia.^2$) (Husch et al., 2003).

2.4.2. Ozone indicator variable

Annual (1999–2003) growing season 4th highest daily max. 8-h average O_3 concentration (modified US and Canadian ambient air quality standard metric form) was calculated from 24 h continuous hourly active monitor data for each elevated O_3 ring. Spatial analysis (ESRI ARC Map; data interpolated using a tension spline, weight 0.1) of 1999–2003 data collected by continuous 24-h monitoring along the Aspen FACE perimeter fence lines showed little intra-season variation in ambient O_3 across the site (Percy, unpublished). Therefore, annual (1999–2003) control ring growing season 4th highest daily max. 8-h average O_3 was calculated from on-site ambient air monitor (Oneida Co, WI: EPA AIRS ID 5508500044420101) data available at http://oaspub.epa.gov/airsdata.

Although the current US and Canadian ambient air quality standard metric form was used to derive the regression models used in this study, three other indices of surface O_3 exposure were investigated as single indicators of aspen mean cross-sectional area growth response in the four O_3 responsive aspen clones (42E, 271, 216, 259): (1) the SUM06 threshold-based sum of all growing season daytime (08:00–19:59 h) O_3 concentrations ≥60 ppb (Lefohn and Foley, 1992); (2) the accumulated over a threshold (AOT)-based sum of all growing season hours of the day with a clear-sky global radiation above 50 W m^{-2} (07:00–21:00 h) O_3 concentrations >40 ppb (Fuhrer et al., 1997); (3) the growing season daily maximum 1-h average O_3 concentration. For this analysis, only data (1999–2003) from the three replicate elevated O_3 rings were used.

2.4.3. Meteorological indicator variables

Annual seasonal meteorological indicator variables were calculated from higher frequency sampling intervals described elsewhere (Dickson et al., 2000) and generally available (www.ncrs.fs.fed.us/4401/focus/face/meteorology). Daytime temperature, photosynthetically active radiation (PAR), wind speed (WS), relative humidity (RH), and precipitation data used in this study were measured at the Aspen FACE meteorological tower. Growing degree days (GDD) or "heat units" were computed by subtracting a base temperature of 10 °C from the average of the maximum and minimum temperatures (5 min scan interval) for each day measured at 10 m. If the daily average temperature computed from the maximum and minimum temperatures was less than 10 °C, the average temperature was set to 10 °C so that the GDD contribution from that day was zero, and not negative. Accumulated growing season PAR (mmol m^{-2} s^{-1}; 5 s scan interval) was calculated as the sum of half-hourly values. Average growing season WS (m s^{-1}; 5 s scan interval; 30 min average reporting) and average growing season 09:00 h RH (%); 5 min scan interval; 30 min reporting) were calculated from data collected at 10 m. Time-specific growing season precipitation (mm) was calculated from monthly sums at the base of the tower. Average growing season soil moisture content (SMC) (%; 2 h scan interval) was calculated from bi-weekly averages taken at 5–35 cm below the surface within the FACE ring aspen communities.

2.5. Statistical analysis

Pearson correlation (Millard and Neerchal, 2001) was used to characterize the relationships between response (cross-sectional area growth) and indicator variables. Multiple regression models (Millard and Neerchal, 2001) were developed using six indicator variables. Although multiple linear regression models of aspen clone growth on O_3 were very highly statistically significant, polynomial cubic regression (Millard and Neerchal, 2001) was investigated in order to evaluate if assumption of non-linearity in tree growth response to O_3 exposure could be verified. To determine the most suitable regression models for assessment of O_3 and the other indicator variables on cross-sectional area growth, the Best Regression Algorithm (Millard and Neerchal, 2001) was systematically applied to each of the five aspen clones. Using this outcome, the best multiple regression models were developed incorporating the three best indicators of aspen cross-sectional area growth. Confidence intervals for the cross-sectional area growth were computed using Monte Carlo techniques (Millard and Neerchal, 2001) to randomly generate thousands of scenarios for all relevant ranges of O_3, GDD, and WS. Resulting confidence (95%) bands in 2-dimensional Euclidean spaces were represented by a graph in a plane for ease of visualization.

3. Results

3.1. Exploratory data analysis

To visualize the range of North American surface-level O_3 concentrations and its recent degree of coincidence with natural populations of *P. tremuloides*, we used the North American ambient air quality standard metric form to map continental O_3 concentrations. The 2001–2003 3-year average 4th highest daily max 8-h average concentrations ranged from 55 ppb to >95 ppb across Canada, the United States, and Mexico (Fig. 1). Ozone concentrations were highest in the Great Lake States, Midwest, east coast, and southern California portions of the US and in southwestern Ontario, Canada.

To place our Aspen FACE O_3 exposure within this North American ambient air quality context, we calculated the 1999–2003 annual growing season 4th highest daily maximum 8-h average O_3 concentrations measured within the three elevated O_3 rings. During the study period 1999–2003, annual growing season 4th highest daily average 8-h maximum O_3 within the elevated O_3 rings ranged from 78 ppb (2000) to 94 ppb (1999) (Fig. 2). There was little ring-to-ring variation within each treatment in annual growing season 4th highest daily max. 8-h average O_3 concentration.

Exploratory analysis with 5-year growth data showed a relationship between decreasing cross-sectional area and increasing (62 to 94 ppb) growing season 4th highest daily max. 8-h average O_3 concentration measured for control and elevated O_3 Aspen FACE ring. This trend was pronounced ($p < 0.02$) for clones 271, 42E, 216, and 259, but not for clone 8L ($p = 0.713$). Average growing season WS was negatively, and highly significantly ($p = 0.000$) correlated with mean cross-sectional area growth over the 5-year period in all five aspen clones. Mean cross-sectional area was negatively correlated ($p < 0.034$) with growing season cumulative PAR in aspen clones 8L, 42E, 216, 259 but not in clone 271 ($p = 0.078$). There was no significant ($p > 0.101$) correlation between mean cross-sectional area and growing season cumulative GDD. Average growing season SMC was positively related with mean cross-sectional area, but not significantly ($p > 0.348$).

We determined the intrinsic relationship between O_3 and tree growth in our data to be non-linear. This can be

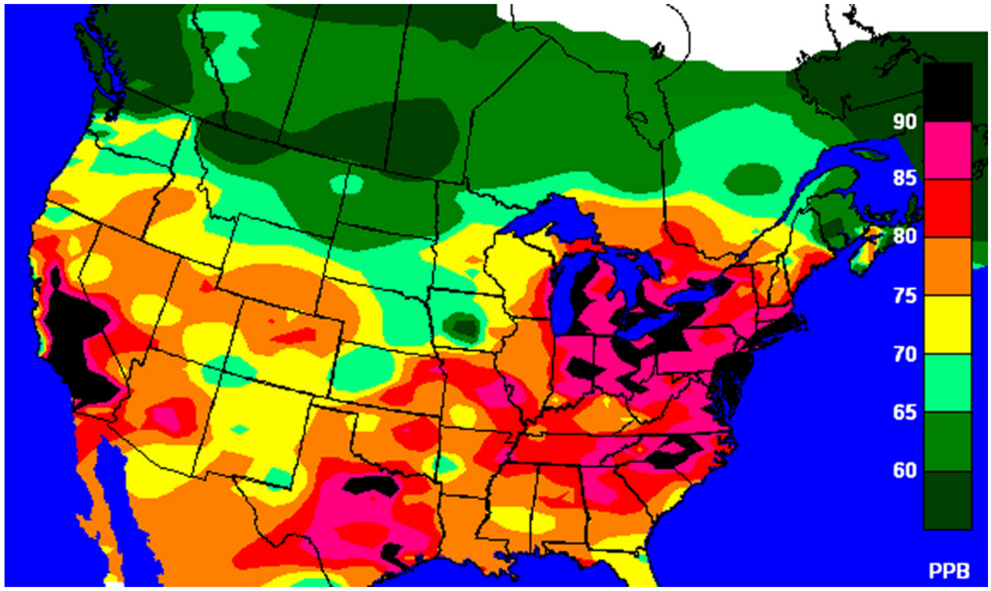

Fig. 1. Spatial distribution of North American surface O_3 calculated using the North American ambient air quality standard metric form 3-year (2001–2003) average of the annual 4th highest daily maximum 8-h average O_3 concentration.

observed from the plots (Fig. 3, clone 216 shown) of cubic regression of tree growth we used to investigate relative performance of several O_3 indices as single indicators of aspen cross-sectional area growth. In the case of the four clones (42E, 216, 271, 259) responding negatively to O_3, growing season 4th highest daily maximum 8-h average O_3 concentration was a much better indicator (higher r^2 adj.; regression ANOVA F-test $p < 0.05$ for clones 42E, 216, 271) of aspen cross-sectional area growth than were SUM06, AOT40, or maximum 1-h concentration (Table 1). This was particularly the case for the more O_3-sensitive aspen clone 42E (r^2 adj. 0.513; $p = 0.012$) as well as to a lesser degree for the more tolerant clone 271 (r^2 adj. 0.454; $p = 0.021$).

3.2. Six-indicator regression models

Complete multiple linear regression models comprising the six indicator variables—growing season 4th highest daily max. 8-h average O_3 concentration, cumulative growing season GDD, average growing season WS, cumulative growing season PAR, cumulative growing season precipitation, and average growing season SMC produced best available fits (r^2 adj. $= 0.687$–0.944) for all five aspen clones. The highest value (r^2 adj. $= 0$ .944; regression ANOVA F-test $p = 0.000$) corresponded to clone 216, which has been shown to be of medium–high sensitivity to O_3. The lowest value (r^2 adj. $= 0.687$; $p = 0.000$) corresponded to clone 8L.

3.3. Three-indicator regression models

To optimize the regression models for risk analysis, we balanced this exceptionally high degree of goodness of fit against the utility requirements of regulators and policy makers by applying the Best Subset Regression Algorithm. Best models (r^2 adj. > 0.650) were those comprising three indicators. Statistical analysis determined the optimum three growth indicators to be growing season 4th highest daily max. 8-h average O_3 concentration, average growing season WS, and cumulative growing season GDD. Soil moisture content proved a useful indicator of growth. However, in regression analysis it was only added to more complex models comprising five indicators. In addition, the incremental goodness of fit added by insertion of SMC was small (e.g., 1.7% to r^2 adj. for clone 216) and the model made more complex.

The convex shape of the cubic response function of tree growth to O_3 with a vertex at approximately 68–70 ppb O_3 (growing season 4th highest daily max. 8-h average) suggests

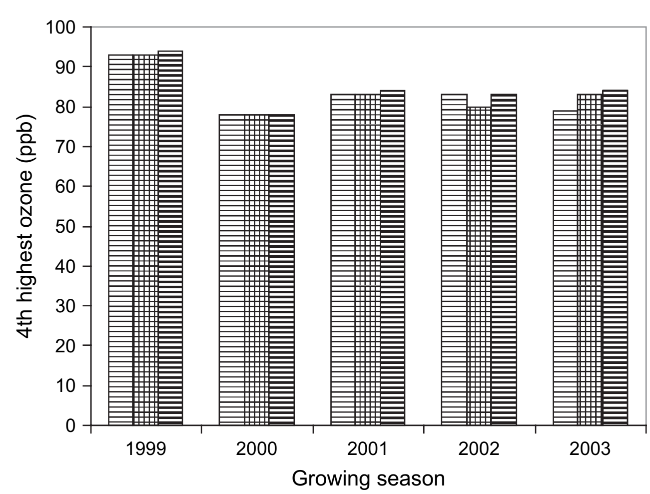

Fig. 2. Growing season exposure in three elevated ozone Aspen FACE rings during 1999–2003. Exposure was calculated as the growing season 4th highest daily maximum 8-h average O_3 concentration measured above the canopy at FACE ring center. Bar groups within each season are the three replicate elevated O_3 FACE rings 1–3, 2–3, and 3–3 respectively from left to right.

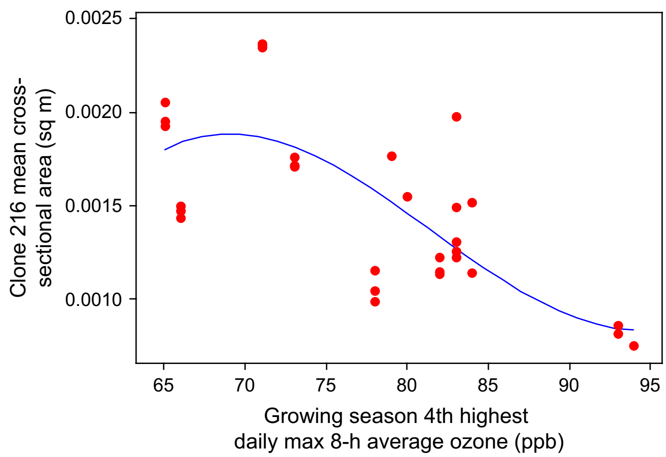

Fig. 3. Cubic regression model for dependence of aspen clone 216 mean cross-sectional area on 1999–2003 growing season 4th highest daily maximum 8-h average O_3 concentration (ppb) fitted using the equation mean cross-sectional area $= 0.06444 + 0.002544$ 4th highest $- 0.000032$ 4th highest$^2 + 0.000000$ 4th highest.

that, whereas O_3 above 70 ppb appears to be clearly detrimental, growth response to concentrations below 68–70 ppb does not seem to be strongly negatively affected (Fig. 3). Although our non-linear (cubic) models could slightly enhance goodness of fit and predictive power, they were less utilitarian. Any increase in predictive power would be very small owing to the extremely high goodness of fit and predictive power achieved by the alternative multiple linear regression model listed below (equation (1) for clone 216).

$$\begin{aligned}&\text{Clone 216 mean annual cross-sectional area } \left(\mathrm{m}^2\right)\\&\quad = 0.00684 - 0.000031\ \text{4th highest } O_3 - 0.00551\ \text{WS}\\&\qquad + 0.000003\ \text{GDD} \qquad (1)\end{aligned}$$

The linear models produced for the five clones are shown in Fig. 4. The models had very high statistical significance (Regression ANOVA F-test $p = 0.0000$), very high goodness of fit (r^2 adj. $= 0.636$–0.882). Considering only our four O_3 responsive aspen clones (271, 42E, 216, 259), the corresponding r^2 adj. ranged from 0.709–0.882 (Table 2). Regression coefficients at growing season 4th highest daily max. 8-h average O_3 were negative and highly significant ($p < 0.038$). Relative contribution of the growing season 4th highest daily maximum 8-h average O_3 concentration was responsible for between 10% and 47.4% of mean tree cross-sectional area growth, depending on relative sensitivity of the clones to O_3. Regression models for clone 8L did not demonstrate any significant negative O_3 effect under the measured meteorological conditions and growing season 4th highest daily max. 8-h average 62–94 ppb O_3.

3.4. Calculation of growth change using regression models

We used the single point prediction technique (midpoint of the prediction confidence interval) derived in our exposure–response functions (black line in Fig. 4) to estimate change in cross-sectional area growth as O_3 concentration increased in 5 ppb increments from a baseline concentration of 60 ppb. Considering a growing season 4th highest daily max. 8-h average O_3 concentration increase from 60 ppb to 80 ppb, cross-sectional area was predicted to increase in aspen clone 8L by 4.3% (Table 3). Clones 259, 271, 42E and 216 all showed decreased growth (−6.9%, −20.8%, −24.3%, and −28.5%, respectively) at 80 ppb O_3 relative to 60 ppb (Table 3, Fig. 4). Potential application of the embedded full confidence interval (upper and lower 95% confidence limits for eventual use to define uncertainty in risk prediction) is described below.

3.5. Calculation of uncertainty

To delineate uncertainty levels in risk prediction application of our models, we randomly generated thousands of plausible scenarios for O_3, GDD, and WS based on actual frequency distributions of these indicators. The 95% confidence bands for prediction of the growing season 4th highest daily max. 8-h average O_3 effects on mean cross-sectional area growth of the five aspen clones (varying sensitivity to O_3) are shown in Fig. 4. Q–Q (quartile) probability plots for the indicators indicated a perfect fit for their distribution. At a given O_3 concentration, a vertical line can be drawn from the x-axis to the intersections with the red, green, and black lines. The black line intersection corresponds to the (single

Table 1
Evaluation of O_3 metric performance as a single indicator of *P. tremuloides* cross-sectional area growth in five clones of contrasting sensitivity

	Aspen clone				
Ozone metric	42E	216	271	259	8L
4th highest[a]	0.513 (0.012)	0.479 (0.017)	0.454 (0.021)	0.179 (0.170)	0.352 (0.078)
SUM06[b]	0.170 (0.180)	0.137 (0.217)	0.163 (0.187)	0.223 (0.130)	0.228 (0.160)
AOT40[c]	0.222 (0.130)	0.190 (0.159)	0.213 (0.138)	0.003 (0.374)	0.375 (0.067)
Max. 1 h[d]	0.250 (0.109)	0.314 (0.069)	0.197 (0.152)	0.121 (0.236)	0.371 (0.069)

Data are r^2 adj. (p values) from cubic regression analysis of dependence of cross-sectional area growth (1999–2003) on growing season 4th highest daily maximum 8-h average O_3 concentration in three replicate elevated O_3 FACE rings.

[a] Growing season 4th highest daily maximum 8-h average O_3 concentration (ppb).

[b] Threshold-based sum of all daytime (08:00–19:59 h) ozone concentration hours ≥60 ppb (Lefohn and Foley, 1992).

[c] Accumulated Over Threshold (AOT)-based sum of all growing season daytime (07:00–20:59 h) ozone concentrations >40 ppb (Fuhrer et al., 1997).

[d] Growing season maximum 1-h average ozone concentration (ppb).

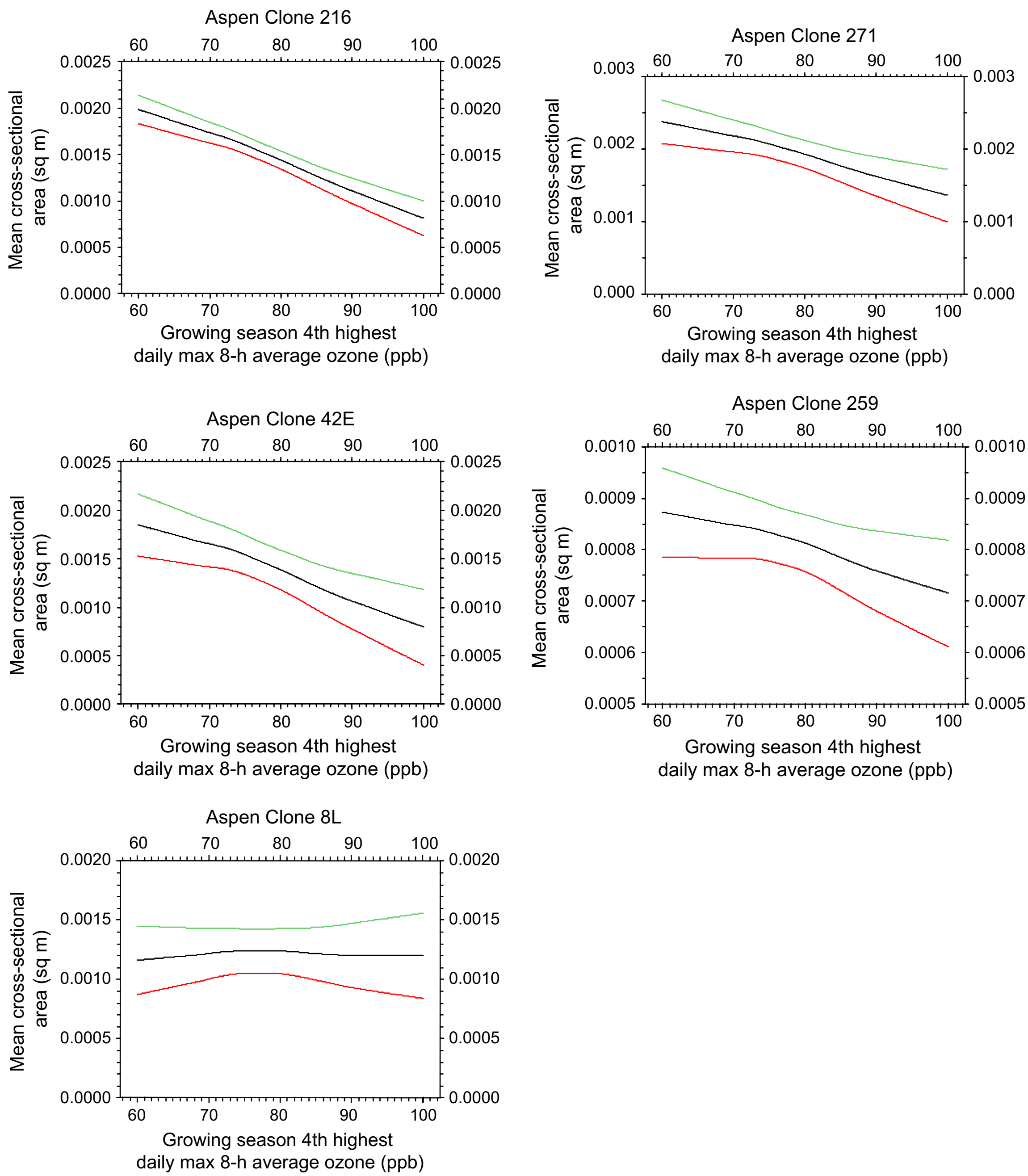

Fig. 4. Exposure–response models (mean ± 95% confidence intervals) for effect of O_3 on mean cross-sectional area growth of aspen clones 216, 271, 42E, 259 and 8L. For any given specific value of the annual growing season 4th highest daily maximum 8-h O_3 concentration (ppb), one can draw a vertical line from that value on the *x*-axis and find the intersections with the red, green, and black lines. The black line intersection corresponds to the (single point) prediction of the average (mean) cross-sectional area response to the given O_3 concentration. This point also serves as a *midpoint* of prediction confidence interval. The range between the red line (lower limit) intersection and the green line (upper limit) intersection corresponds to the 95% confidence interval (95% CI) for mean cross-sectional area response. Grid lines have been removed for clarity of presentation.

midpoint) prediction of the average (mean) cross-sectional area response to the given value of growing season 4th highest daily max. 8-h average O_3 concentration. The accuracy of the forecast (the width of the corresponding confidence) depends on the actual growing season 4th highest daily max. 8-h average O_3 concentration. Confidence bands (Fig. 4) embedded within the midpoint predictions can be used to meet a critical requirement by regulators for defining uncertainty in risk prediction.

3.6. Application of regression models within the ambient air context

To apply our exposure–response functions at the landscape level within the North American air quality context, we merged the digital representation (http://esp.cr.usgs.gov/data/atlas/little) of aspen distribution with the 3-year average of the annual 4th highest daily max. 8-h average O_3 concentration (Fig. 1) as mapped using EPA AIRS software. In order

Table 2
Summary of multiple regression model of aspen clone mean cross-sectional area (m^2) on growing season 4th highest daily maximum 8-h average O_3 concentration, average growing season WS and cumulative growing season GDD

Aspen clone	Model significance	4th highest O_3 effect	4th highest O_3 significance	r^2	r^2 adj.	4th highest O_3 contribution
8L	$p = 0.0000$	Negative	$p = 0.900$	0.676	0.636	6.8%
259	$p = 0.0000$	Negative	$p = 0.038$	0.739	0.709	17.8%
271	$p = 0.0000$	Negative	$p = 0.001$	0.757	0.729	28.6%
42E	$p = 0.0000$	Negative	$p = 0.001$	0.762	0.734	10.0%
216	$p = 0.0000$	Negative	$p = 0.000$	0.894	0.882	47.4%

to produce a representative forecast for the natural population that did not predispose toward either the best (clone 8L) or worst (clone 42E) case scenarios, we averaged responses across the five clones that represented a wide range of *P. tremuloides* sensitivity to O_3 (Table 3). In areas of its range (Midwestern and Great Lake States, parts of California), 3-year average 4th highest daily max. 8-h average O_3 concentration reached 95 ppb and *P. tremuloides* growth loss was predicted to have been −31% (Fig. 5). Over most of western and northern Canada (except for northwestern Alberta, −6%) and parts of the intermountain states, *P. tremuloides* was predicted to have experienced no growth change, or a growth loss of −3%. All the *P. tremuloides* range in the Great Lake States, southern to mid Ontario, Quebec, and southwestern Nova Scotia was predicted to have had growth loss between −11% and −25%. Populations of *P. tremuloides* in Mexico, Arizona, Colorado, and Utah were predicted to have experienced growth loss in the 15% to 25% range (Fig. 5).

4. Discussion

The interaction of O_3 with trees is a very complex process that varies in response to a host of environmental (Kubiske et al., 2006), pest (Percy et al., 2002), and other factors. This presents large challenges when scaling impacts beyond the tree level (Samuelson and Kelly, 2001). Earlier, Grünhage and Jäger (1994) suggested that O_3 flux into the plant could be used as an underlying mechanism for establishing standards to protect vegetation. This was partly due to an acknowledgement (US EPA, 1996) that exposure indices do not completely characterize potential for O_3 uptake, its detoxification or, biochemical interaction within the plant; nor do they characterize the suite of physical, biological, and meteorological processes influencing O_3 deposition and transfer predicting vegetation response. In short, establishing cause−effect relationships for ambient O_3 exposure and tree growth has proved to be an elusive goal (Manning, 2005a).

Table 3
Change in aspen cross-sectional area growth from 60 ppb to 80 ppb growing season 4th highest daily maximum 8-h average O_3 as calculated using exposure−response regression models

Aspen clone	Growing season 4th highest daily maximum 8-h average O_3 (ppb)			
	65	70	75	80
216	−7.0	−13.5	−24.5	−28.5
259	−1.2	−3.4	−4.6	−6.9
271	−4.2	−8.3	−12.5	−20.8
42E	−5.4	−10.8	−16.2	−24.3
8L	+2.5	+3.4	+4.3	+4.3
Population average[a]	−3.03	−6.54	−10.71	−15.26

[a] Average calculated with equal weighting assigned to each clone.

Prior to the availability of O_3 response data on mature trees grown in free-air settings, Hogsett et al. (1997) used seedling response data from mainly chambered experiments and a modified SUM06 O_3 exposure index to characterize risk to eastern forests. The potential for improved O_3 risk prediction through modeling has been enhanced recently through the use of multifactor dendroecological approaches (McLaughlin and Downing, 1995, 1996; McLaughlin and Nosal, in press) and by the advent of long-term free-air exposure experiments with mature trees (Karnosky et al., 2005; Karnosky et al., accepted for publication). Like the dendroecological approach, open or "free-air" O_3 exposure systems enable the investigation of larger, mature trees growing at the stand level under realistic competition and inter-annual variation in physical climate. Therefore, within the North American context of air quality regulation, the potential use of an exposure-based metric, such as that proposed for human health (Foley et al., 2003), to establish surrogates for realistic tree flux−effect relationships (Grünhage et al., 2004) can clearly be enhanced through the use of data derived from free-air experiments established in the ambient air context.

Krupa et al. (2003) postulated that "…it should be possible to build an appropriate and inclusive predictive model comprising all important meteorological indicators plus soil moisture data that, together, would yield a first-order approximation of atmospheric O_3 flux and stomatal uptake." Building on their approach, we used indicator and response data recently available from a long-term, ecosystem-scale free-air manipulative experiment (Karnosky et al., 2005) in which air quality and meteorological measurements were coupled in time and space against a backdrop of inter-annual variability in climate (Kubiske et al., 2006). To partly address some of the scaling challenges enunciated by Samuelson and Kelly (2001) and to advance forest O_3 risk analysis within the North American ambient air quality context, our initial exposure−response regression analysis was completed using models comprising key meteorological indicators (GDD, WS, PAR, RH, precip.) of O_3 concentration and one important O_3 flux regulator (SMC) that would have been expected to yield a first-order approximation of atmospheric O_3 flux and stomatal uptake, as originally proposed by Krupa et al. (2003).

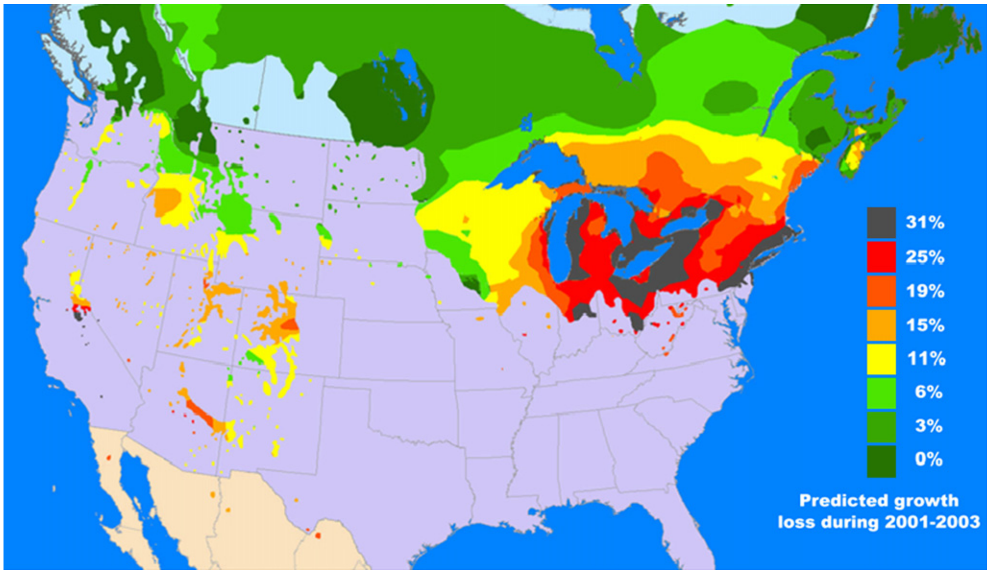

Fig. 5. Estimation of trembling aspen growth loss across North America due to 3-year (2001–2003) average of the ambient annual 4th highest daily maximum 8-h average O_3 concentration. The digital representation (reproduced from Little, 1971) of the natural trembling aspen range (http://esp.cr.usgs.gov/data/atlas/little) was integrated with the surface O_3 distribution in Fig. 1. Change in growth was calculated using the new exposure–response models and then averaged across the four responsive and one unresponsive aspen clones to yield an average species estimate representative of a wide range of O_3 sensitivity. This estimate was finally applied to the integrated spatial map. 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 due to unavailability of O_3 monitoring.

The response endpoint incorporated in our exposure–response models was tree diameter, later converted to stem cross-sectional area. During the growth period covered by this study (1999–2003) average aspen height in the control rings increased from 2.8 to 5.8 m and canopy closure was completed by 2002. As Hogsett et al. (1997) stated, for utility in prediction, the species-level assessment endpoint chosen must have social, economic, and ecological relevance. Productivity, as assessed through diameter, certainly meets this requirement. As diameter could not measured at 1.3 m above ground throughout the study period (see Kubiske et al., 2006) we did not feel justified in scaling our diameters to the forest inventory standard measure of density, namely, basal area (m^2 ha^{-1}) (Husch et al., 2003).

The regression r^2 adj. obtained in our initial six indicator exposure–response models ranged from 0.62 to 0.95, and explained an extremely large proportion of variability in the data in terms of O_3, meteorological and one flux-related indicators. To measurably enhance model utility (ease of use by regulators) while maintaining a high degree of fit and predictive power, we next optimized our predictive models around annual growing season 4th highest daily max. 8-h average O_3 concentration and two important meteorological indicators (GDD, WS) of surface O_3 (National Research Council, 1991) concentration. As correlation analysis showed that RH and precipitation were very highly inter-correlated ($r = 0.799$; $p = 0.0000$) and co-linear with respect to other indicators, RH was omitted as a indicator variable in our final regression analyses. In our case, as previously described, average seasonal WS was chosen as the third indicator in our model. There is no question that soil moisture status is important in regulating O_3 flux into a plant. However, wind speed performed equally as well as SMC. Secondly, it also has the advantage of being reported continuously over the landscape, thus, contributing to greater model utility and perhaps making a contribution to increased scientific literacy (Orbach, 2005).

Many models used previously to predict forest productivity change due to O_3 (Ollinger et al., 2002; Felzer et al., 2004) have assumed a degree of linearity in response to O_3 exposure. Here, using non-linear cubic regression of tree growth on O_3 (Fig. 3), we have in fact demonstrated that: (1) tree growth response to O_3 exposure is intrinsically non-linear; and (2) that this non-linearity is statistically significant. This fact has not been previously widely reported in the literature! As explained above, for model utility we ultimately opted for linear exposure–response models in which O_3 regression coefficients by themselves contributed between 10% and 47% (Table 2). This compares favorably with the O_3 regression coefficient (other indicators were temperature, VPD, radiation and precipitation) contribution (32% and 40%) to overall model significance calculated by McLaughlin and Nosal (in press) using the dendroecological approach. To improve understanding of O_3 exposure, natural system response and risk, Laurence and Andersen (2003) concluded that one of the five objectives for future research should be to “build mechanistic models that quantify and propagate uncertainty so that we may provide a useful interpretation of our science for those in the policy arena.” Our new exposure–response models are able to predict within the broad range of recent North American continental-scale 3-year average of the annual 4th highest daily maximum 8-h average O_3 concentrations (Fig. 1). This is not surprising as the O_3 indicator range (growing season 4th highest daily maximum 8-h average O_3; 62 ppb ambient to 94 ppb fumigated) experienced during 1999–2003 at Aspen

FACE has immediate relevancy to ambient O_3 concentrations in the United States, Canada, and Mexico.

Using the three-indicator exposure–response models, we estimated growth change between latitudes 24° and 56°, or approximately two thirds of the latitude over which *P. tremuloides* grows. Unfortunately, a lack of surface-level O_3 monitoring in northern Canada and in Alaska precluded extending our estimates further north. Growth change calculated using the clonal models across the spatial distribution of 2001–2003 O_3, then averaged to yield a population estimate, produced retrospective estimates of growth offsets between 0% and -31% (Fig. 5). Over the majority of the natural aspen range in north central, upper mid-western and northeastern trans-boundary area, growth was predicted to have decreased −11% to −25%. Hogsett et al. (1997) predicted that the area-weighted response of annual aspen seedling biomass from historical (1988–1989) O_3 exposure calculated using a modified SUM06 index was −14% to −33% over 50% of the eastern United States. Moderately sensitive species *Liriodendron tulipifera, P. taeda, Pinus strobus*, and *A. saccharum* were predicted to have had a −5% to −13% biomass loss. Our models were based on annual measures of indicators and response to derive a prediction of the cumulative O_3 effect. McLaughlin et al. (2003) measured diurnal changes in physiologically important responses over two seasons of contrasting O_3 exposure, that O_3 provides a significant contribution to day-to-day variability in *L. tulipifera* stem growth. They also raised the possibility that cumulative effects have the potential to be important over multiple years of high O_3 exposure. Our modeled estimates also lie well within the range of those published (up to 50% in high O_3 years) for other hardwood tree species by McLaughlin and Nosal (in press).

As with any fumigation technique used over the past 50 years to investigate plant response to O_3 (see review by Karnosky et al., in press), the level of O_3 control in a free-air experiment such as Aspen FACE must be considered when interpreting results. The treatment record (www.aspenface.mtu.edu) indicates that the FACE system performance has been consistently very good. At Aspen FACE, O_3 concentrations were continuously monitored at ring center/canopy height and then applied to the aspen populations. Since 2003, we have deployed growing season arrays (12 locations per ring, two heights) of CANOXY passive O_3 samplers (Cox and Malcolm, 1999) within the O_3 FACE rings in order to: (1) describe the monthly vertical/horizontal distribution of O_3 exposure within FACE rings; and (2) potentially use these cumulative O_3 exposure data to mimic (Krupa and Nosal, 2001) corresponding frequency distributions of hourly O_3. Recent spatial analysis for the 2005 growing season indicates that the monthly horizontal and vertical distribution of surface O_3 exposure is relatively good within the FACE ring core area where the response measurements were taken (Karnosky et al., accepted for publication). For instance, at the ring center where the O_3 inlet for continuous active monitoring and treatment control is situated, there were no significant differences between 2005 monthly SUM00 measured at 10-m, 4-m and 2-m heights using passive sampling techniques. September SUM00 O_3 was higher than the June–August (Karnosky et al., accepted for publication) likely due to O_3 induced early leaf abscission (Karnosky et al., 2005). Ultimately, however, reducing further uncertainty around O_3-distribution within FACE rings requires an intensive campaign of continuous multi-port sampling co-located with passive monitors.

Based on much earlier examination of: (1) foliar symptoms on some 220 aspen clones representing 15 populations distributed across the natural range of aspen in the conterminous United States (Berrang et al., 1986); and (2) the subsequent growth and biomass responses of these clones planted in the Lake States region (Berrang et al., 1989; Karnosky et al., 1996), we believe that the five clones used in this study represented a very wide range of O_3 sensitivity that might be expected to occur within natural populations. Genotypic variation in response to O_3 among the clones (Karnosky et al., 1996, 2003b) studied is not surprising and is a well-known phenomenon in most species. In this analysis, four clones (271, 42E, 216, 259) of different sensitivities to O_3 responded negatively to increasing O_3 concentrations. Clone 8L did not respond strongly negatively to O_3 between growing season 4th highest daily max. 8-h average O_3 concentrations between 60 ppb and 90 ppb. Aspen clone 8L is known to be extremely O_3 tolerant (Karnosky et al., 2005). However, it is possible that clone 8L might respond negatively to higher O_3 concentrations that occurred during 2001–2003 within the Southwest/Midwest/central-mountain, mid-eastern US, southern Ontario/Quebec/Nova Scotia in Canada, and within *P. tremuloides* populations in Mexico (Fig. 1).

By averaging clonal response within the context of a species-level risk analysis, we believe that our data describe the intrinsic relationship between aspen growth, O_3, and two key meteorological indicators. To increase our level of confidence, we are further examining the longer-term (10 years) growth response for a set of genetic materials (clones 271, 216, 259) common to those planted at Aspen FACE, but growing in plantations at several locations in the Great Lakes States. We expect that, based upon earlier measurements (Karnosky et al., 1999, 2003a), our new metric-based exposure–response models will be validated against growth change observed within the range of uncertainty inherent in the models. Along with Hogsett et al. (1997), we of course recognize that modeled estimates of growth change due to O_3 derived from aspen monocultures at Aspen FACE and our field plantations may be modified to a degree across the forest landscape as more O_3-sensitive aspen clones may not compete as well when growing with other more O_3-tolerant species like *B. papyrifera* (McDonald et al., 2002; King et al., 2005).

Like any other manipulative technique used previously (see Karnosky et al., in press) for exposing trees to O_3, free-air exposure systems like Aspen FACE are limited in their utility of investigation of tree response under sufficiently wide range of O_3 exposure scenarios normally required for investigations using regression methodology. For more rigorous model building, O_3 exposures from 50 ppb to 120 ppb would be required. Of course, this is practically impossible (or at least extremely expensive) in a free-air system, and most other approaches, except perhaps in the case of the dendroecological approach of McLaughlin and Nosal (in press). However, as O_3 exposure is not controlled, the dendroecological approach also has its limitations when trying to relate response to exposure over time and space, and typically requires a more complex and less utilitarian modeling approach.

In a comprehensive retrospective review of the roles of air pollutants and climate in North American forest health, McLaughlin and Percy (1999) reported that O_3 was deleteriously affecting forest ecosystem function across large and geographically widely separated areas of the continent. When considering relevance of this current research within the ambient exposure-based metric context (Foley et al., 2003), it is important to note that the growing season 4th highest daily max. 8-h average O_3 concentration indicator used in our models in fact represents the *biologically relevant* portion of the NAAQS (Federal Register, 1997) and CWS (CCME, 2000). It encompasses the period (stage 3 of bud break to leaf fall) during which active O_3 uptake by northern temperate hardwood trees would be expected to occur. Significantly, at almost all sites in North America, the 4th highest O_3 value occurs during the May–August period covered by our Aspen FACE growing season 4th highest daily maximum 8-h average O_3 concentrations.

The exposure–response metric-based growth loss estimates mapped within the 2001–2003 North American ambient air context (Fig. 5) were derived by averaging across a wide range of clonal sensitivity. Uncertainty around the midpoint predictions was then calculated (Monte Carlo analysis) using a wide and very real and plausible range of ambient values for the three indicators O_3, GDD and WS. Delineation of uncertainty is a key requirement and it is the inherent uncertainty in a parameter (95% confidence intervals) combined with its potential importance in a process (diameter growth) that, according to Laurence and Andersen (2003), confers its impact estimation of risk. Our regression analysis indicated that the use of within-ring meteorology conferred an insignificant additional goodness of fit when compared with site-level data collected at the meteorological tower. Therefore, meteorological data collected at a central location (meteorological tower) were used to derive regression predictor values, thus, further increasing the potential for model application across the landscape. Hitherto, the usefulness of exposure-based metrics to assess risk has been questionable, particularly for forest ecosystems subjected to seasonal droughts (Panek et al., 2002).

The cubic curves of tree cross-sectional area growth response to O_3 displayed a significant degree of curvature and a significant improvement in goodness of fit when compared with a simple linear model. One observation that can be made from our cubic regression models (clone 216, Fig. 3) is that the response surface had a convex shape with a vertex at approximately 68–70 ppb. The suggestion that O_3 above 70 ppb appeared to be detrimental, whereas concentrations below 68–70 ppb did not appear to be detrimental is interesting. In the science of O_3 forest response, investigators have too often placed emphasis on finding only strongly negative responses (Manning, 2005b). Calabrese (2005) has convincingly stated the case for the hormetic dose–response relationship as underlying the toxicological basis for risk assessment. Our cubic regression curves are certainly compatible with the concept of hormesis. However, in order to demonstrate hormesis, we would need many more data points with O_3 concentrations ranging well below and above the values that we had available, particularly given the variability in tree response at any given O_3 concentration. Therefore, further modeling with a much wider range of O_3 concentrations will be needed to reliably confirm this perceived compatibility.

With three (42E, 216, 271) of our four O_3 responsive clones exposed to elevated O_3, the growing season 4th highest daily maximum 8-h average O_3 concentration metric was determined to perform better (higher r^2 adj.; regression ANOVA F-test $p < 0.05$) than growing season SUM06, AOT40, or max. 1-h average O_3 concentration as a single indicator of aspen cross-sectional area growth (Table 1). Cubic regression analysis of the performance of four O_3 metrics indicated negative and statistically significant ($p < 0.05$) relationships between growing season 4th highest daily maximum 8-h average O_3 concentration and growth in three aspen clones. Although these results might be of particular interest to regulators, it should be noted that the analysis was completed using only data collected in the three elevated O_3 rings where O_3 was continuously monitored. In other words, the distribution frequency would have been skewed toward higher average hourly concentrations and greater accumulated exposure than if data from the three control rings had also been included. Therefore, in our view these results are indicative only of relative metric performance, and do not imply that growing season 4th highest daily maximum 8-h average O_3 concentration can be used by itself as an indicator of aspen growth.

5. Conclusions

Greater demands are being placed on scientists for sound, biologically relevant, exposure-response research that can be applied within an ambient air management context. Exposure-based O_3 metrics are the only practical method of relating air quality standards to vegetation response within the North American ambient air context at this time. Accordingly, we have developed new exposure-based response relationships designed to reflect the ambient air quality reality in North America. The regression models comprising growing season 4th highest daily maximum 8-h average O_3 concentration, growing degree days, and growing season average wind speed: (1) were highly statistically significant; (2) had a high degree of goodness of fit; (3) included defined levels of inherent uncertainty for an important endpoint; (4) were biologically relevant; and, (5) should be simple to use within the North American context. Historical 2001–2003 growth change due to ambient O_3 over most of the 26 M ha *P. tremuloides* distribution was estimated to have ranged from no impact (0%) to strong negative impacts (−31%). The models have potential for use in parallel with those proposed for the human population to estimate cost–benefit implications of various O_3

precursor control strategy scenarios for achieving exposure reduction.

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