A review of statistical methods for the meteorological adjustment of tropospheric ozone

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ABSTRACT:

A variety of statistical methods for meteorological adjustment of ozone have been proposed in the literature over the last decade or so. These can be broadly classified into regression methods, extreme value methods, and space-time methods. We describe and offer a critical review of the approaches, discuss questions of variable selection and trend estimation, and compare selected methods as applied to ozone time series from the Chicago area.

Key Words: Regression, extreme values, time series, spatial statistics, environmetrics.

1. Introduction

The meteorological adjustment of tropospheric ozone can be achieved by statistical modeling of the association between ozone concentration and meteorological variables. The last decade has seen a growing diversity of statistical literature on the subject with the application of a wide range of statistical methodologies, the use of widely differing data, and with adjustment being considered for different policy objectives. This critical review provides an evaluation of this literature focusing on differences in model structure. We categorize models under three broad statistical approaches: regression-based modeling, extreme value approaches, and space-time models. We also compare selected methods applied to the derivation of a meteorologically adjusted ozone series and subsequent investigation of time trend using data from the Chicago area.

The main objectives for meteorological adjustment of surface ozone measurements include: (a) obtaining air quality forecasts, (b) investigating and estimating ozone time trends, and (c) increasing scientific understanding of the underlying mechanisms. The objective can influence the choice of both appropriate statistical methods and relevant data. Forecasting extreme ozone events in order to provide public health warnings may focus the analysis on investigating those observations exceeding a threshold and their association with readily predicted meteorology. In contrast, assessing time trends may involve modeling all available daily surface ozone observations and their relationship to any reasonable and available surface or upper air meteorological covariates. Models developed to provide insight into the conditions conducive to ozone production may present additional data demands for measurements of precursors as well as synoptic-scale meteorology. This is the focus of photochemical modeling research, but has not received much attention in the statistical literature.

A further context for assessing the relationship between ozone and meteorology, which we do not specifically consider, is the analysis of ozone and meteorology jointly in the determination of potential ozone-related health effects (Stieb et al., 1996, Kuenzli et al., 1997). The statistical issues in the analysis of health effects are different from those considered here because ozone is a predictor rather than an outcome in the modeling and the impact of meteorology would be considered in the context of epidemiological confounding, such as temperature affecting both ozone production and hospital admissions.

2. Data Considerations

Even within the context of a particular modeling objective, the data used in the literature vary widely both in terms of the variables considered (ozone summaries chosen, surface and/or upper air meteorological variables included) and in terms of observation scales in space (single monitor, network of monitors) and time (hourly, daily, seasonal, annual). Different sets of meteorological variables are used depending on local and synoptic meteorology and data availability. We briefly summarize the temporal and spatial scales of ozone observations commonly used, as well as the meteorological variables investigated and eventually incorporated into each analysis.

***Table 1 about here ***

2.1 Ozone measures

2.1.1 Time scales

The time scales for summary measures of ozone range from 5-minute to daily summaries, most commonly daily 1 hour maximum levels (Table 1). Daily maximum 1-hour and maximum 8-hour averages are the focus of the current U.S. National Ambient Air Quality Standards (EPA, 1998a) because of expected health effects of exceedances. Daily summaries are also the basis of most assessments of trend. Finer (hourly) scales are most relevant for process modeling, short-term predictions, and photochemical model evaluation. Daily median and 90th percentile have also been considered. Ozone data are most often modeled in terms of original concentration scales (parts per million), although in some cases transformations, such as square root or logarithm, are used. The appropriate transformation will depend on the temporal and spatial scale of the particular analysis, with greater spatial and temporal averaging domains generally resulting in more nearly Gaussian distributions.

2.1.2 Spatial scales

There is also considerable variety in the spatial scales represented in analyses of ozone measurements (Table 1). While several studies use data from a single location, many consider a regional network of ozone monitors. Analyzing data from a regional network requires a decision on how to handle the spatial ozone field. Among the ozone network analyses reviewed here, most model each monitor independently, some model a derived univariate network summary, and one models the multivariate spatial field.

Separate modeling of the association between each ozone monitor and local meteorology is the simplest and most common approach to analysis. However, this approach ignores any information on regional dynamics of meteorology and ozone available in the analysis of a network of ozone monitoring sites, and may therefore result in a statistically less powerful or possibly misleading analysis for purposes such as the assessment of regional trend. Analysis of the full network response through space-time modeling resides at the other end of the spectrum, being more complex both theoretically and computationally, yet potentially more flexible in its ability to capture regional associations between ozone and meteorology. Modeling a univariate summary of the ozone network retains the simplicity and wealth of tools available for univariate responses, but requires choosing the appropriate summary.

Two broad solutions to the question of choice of spatial summary appear in the literature. The first involves selecting a simple network summary. Examples include network average daily 1 hour maximum (Stoeckenius and Hudischewskyj, 1990; Eder et al., 1994) and the network maximum of the daily 1 hour maxima, (Niu, 1996; Smith and Huang, 1993; Stoeckenius and Hudischewskyj, 1990). Alternatively, network summaries have been derived using multivariate dimension reduction techniques. For instance, principal components analysis of a network of ozone monitors provided a univariate network summary for the modeling of Bloomfield et al. (1993a, 1993b), and the subsequent reanalysis by Gao et al. (1994).

A contrasting approach in this same context uses the singular value decomposition (SVD) of the cross-covariance matrix between measures from a network of ozone monitors and a network of meteorological monitors to derive the univariate ozone and meteorology summaries that are most highly associated in terms of explained covariance (Reynolds et al.,1998). In application of these methods to data from the Chicago region, the ozone network summaries were approximately proportional to network averages, allowing simple interpretations of the analysis (see section 4).

The modeling of ozone network data requires a balance between interpretation, simplicity of approach, and incorporation of regional-scale response information for increased statistical power. Multivariate reduction techniques seem a promising middle-ground for trend investigations, provided the resulting summaries are scientifically interpretable.

2.2 Meteorological data

2.2.1 Ozone Chemistry

The fundamental production vehicle for ozone is photochemistry. A substantial proportion of anthropomorphic ozone production obtains from the sequence of reactions

$$NO_2 + h\nu \rightarrow NO + O$$

$$NO + O_3 \rightarrow NO_2 + O_2$$

$$O + O_2 \rightarrow O_3$$

the first reaction being photochemical. In addition, similar reactions with volatile organic compounds (VOCs) are important in the formation of ozone. Generally speaking, conditions favorable to ozone production are sunshine, high temperatures, and stagnant air (NRC, 1991).

2.2.2 Relevant meteorological variables

The choice of relevant meteorological variables depends on the purpose of the analysis, regional differences in meteorology and emission patterns, and data availability. Forecasting restricts the analysis to readily predicted (or lagged) meteorology and, in the context of forecasting extreme events, focuses on just those meteorological variables indicative of such events. Alternatively, assessing trends and other long-term developments necessitates study of ozone production under a variety of atmospheric conditions. Meteorological variables considered and included in the reviewed literature are summarized in Table 2. For each application, we list only those variables that are explicitly mentioned in the corresponding paper.

***Table 2 about here ***

Cox and Chu (1993, 1996) considered some 100 meteorological variables and found maximum surface temperature, wind speed, relative humidity, mixing height, and opaque cloud cover as well as wind speed $\times$ temperature interaction, to be significant meteorological predictors over most major metropolitan areas of the United States. However, regional differences in meteorology and emissions patterns make more precise statements of dominant variables tenuous. Other studies have reached similar conclusions.

Upper air measurements have been included in a number of studies. They appear most beneficial for predicting the conditions underlying extreme events (Burrows et al., 1995; Pryor et al., 1995) as such events have been shown to be associated with specific synoptic weather patterns (e.g., Eder et al., 1994; McKendry, 1994). A source of noise in the observed upper air–ozone relationship is that upper air measurements are generally only available at the hours of 0000 and 1200 UTC, translating to USA local standard times of 4–7 pm and am, respectively.

2.2.3 Spatial scales

Another issue concerns the relative locations of the ozone and meteorology monitors. While airports often provide high quality meteorological surface and upper air observations, measurements from monitors which are not co-located with the air quality monitors are frequently used, as well as analyzed regional meteorological data resulting from a variety of sources including radar, satellites and balloon measurements. Analyses that we have carried out in Washington State (Reynolds et al., 1998) and Chicago (section 4) indicate that airport meteorological data may provide the most reliable meteorological measures for associations with ozone over broad regions.

3. Methods

The statistical approaches to ozone adjustment for meteorology may be divided into three broad areas, with considerable variety within each: regression-based modeling, extreme value approaches, and space-time models. A common focus is the reduction of unexplained variability in ozone through meteorological adjustment. Rao et al. (1997) argue that process changes due to policy or climate changes may be very small and difficult to detect unless they are separated from weather and seasonality. Regression-based and extreme value methods are aimed primarily at forecasting or trend estimation, and to a lesser degree at elucidating underlying mechanisms. Space-time modeling has so far received little attention in the literature but could, in principle, address all three adjustment goals, with the disadvantage of increased complexity of modeling and data collection.

3.1 Regression based methods

The majority of approaches to meteorological adjustment of ozone are in some sense regression-based, with widely varying degrees of complexity. The structure of these models can be considered in three broad categories: linear regression, regression trees, and non-linear regression. Within each category there are further distinctions such as the method of introducing meteorological variables (directly or via dimension reduction) and the incorporation of temporal and spatial dependence. The class of regression models dictates the availability of software for model fitting and diagnostics, the ability to model complex interactions among the meteorological variables, and the familiarity and ease of interpretation for the policy-maker.

3.1.1 Models based on linear regression

The most familiar of the methodologies employed in the literature is linear regression. In its simplest form (e.g. Korsog and Wolff, 1991; Feister and Balzer, 1991; Abdul-Wahab et al., 1996; Katsoulis, 1996; Fiore et al., 1998), multiple linear regression modeling is used to link ozone measurements to contemporaneous meteorological measures. The absence of consideration of lagged auto- and cross-correlation makes the scientific merit of such models questionable. Next in complexity is time series modeling which incorporates lagged relationships and a correlation structure (usually a simple AR(1)) for the residuals (e.g. Galbally et al., 1986). Provided that fits are confirmed by suitable diagnostics (see Section 3.5), this approach may provide reasonably robust models for forecasting. However, the simple linear associations between the variables are unlikely to aid understanding of processes driving the relationships, in that they may be inadequate to capture interactions and non-linearities in the ozone response. In addition, incorporation of lagged ozone measurements is not suited to the estimation of time trends.

Another approach, still within the framework of multiple linear regression, involves models based on linear combinations of meteorological variables, such as factor analysis (Spichtinger et al., 1996), varimax principal component analysis (Pryor et al.,1995; Poissant et al., 1996), and singular value decomposition or canonical covariance analysis (Reynolds et al., 1998). The various methods for dimension reduction enable handling large numbers of potentially multicollinear covariates, reducing the complexity of possible interactions to be investigated. However, they may overlook nonlinear associations. The same concern arises in forming ozone network summaries (see section 2.1.2).

Finally, there are approaches which involve some filtering of both ozone and meteorological variables before the resulting filtered variables are modeled by linear regression (Flaum et al., 1996; Porter et al., 1996; Eskridge et al., 1997; Rao et al., 1997; Milanchus et al., 1998). Details are provided in section 3.3.

3.1.2 Models based on regression trees

Classification and Regression Trees (CART; Breiman et al., 1984) have been explored for their ability to capture nonlinearities in the relationship between meteorological variables and surface ozone with a simpler methodology than construction of a non-linear regression model (Stoeckenius and Hudischewskyj, 1990; Burrows et al., 1995; Huang and Smith, 1999). CART has performed fairly well in forecasting high ozone events, capturing the meteorological interactions most commonly associated with these extreme events (Burrows et al., 1995; Huang and Smith, 1999). However, a trend component cannot be directly incorporated into the tree model.

Huang and Smith (1999) separately fit linear trend models to the observations in each terminal node of their tree, i.e., they investigate trends separately at each cluster of meteorological conditions identified by CART. They find that meteorological clusters conducive to higher ozone tend to display stronger (downward) temporal trends. Their analysis thus provides insight into the interaction between time trends and different meteorological conditions.

Alternatively, Stoeckenius and Hudischewskyj (1990) use a tree model and re-estimate its coefficients for each year using a moving window of data (keeping the model structure fixed). These year-specific models are then used to estimate annual expected numbers of exceedances under climatology as observed over the period of the analysis, which are then assessed for temporal trend. The approach is statistically problematic, due to the varying precision with which the exceedance probabilities are estimated, both across terminal nodes of the tree and across years.

3.1.3 Models based on non-linear regression

Bloomfield et al. (1996) argue that statistical linear models "have difficulty capturing the complex relationships between the meteorological variables and ozone." They develop a parametric non-linear model for data from 45 monitoring stations in the Chicago region over 1981-1991 represented in the AIRS data base (EPA, 1998b). These authors model the daily median (across sites) of the daily site-specific maximum 1-hour average ozone values, using nonlinear least squares. Parametric forms for trend and the relationship between contemporaneous and lagged ozone, surface temperature, relative humidity, surface wind speed and 700 hPa wind speed are identified in stages by exploratory graphical displays and non-parametric modeling. Seasonal terms are modeled via a short Fourier series. In estimating the standard errors of the fitted coefficients, the authors acknowledge the existence of serial autocorrelation in the model residuals and make appropriate adjustments, using the methods of Gallant (1987).

The exploratory multivariate graphical displays used have the advantage of revealing interactions in the relationships between the various meteorological variables in their association with ozone. For instance, the scale of the ozone association with temperature depends on relative humidity. The displays further allow flexibility in motivating non-linear functional forms for the ozone model.

Davis et al. (1998) consider the same form of ozone network summary used by Bloomfield et al. (1996) for 11 sites in the Houston, Texas area and meteorological variables from a single site for the period 1981-1992. They use cluster analysis applied to daily meteorological component scores (obtained from SVD) to separate the records into 7 clusters, which show significantly different ozone concentrations. The relationship between ozone and meteorological variables is then modeled separately in each cluster using stepwise selection and generalized additive models (Hastie and Tibshirani, 1990). The decomposition of the record into clusters is similar in spirit to Huang and Smith (1999), except that Davis et al. then also model the association of ozone with meteorology within each cluster. It is not clear how seasonality and autocorrelation are accommodated in the modeling.

3.2 Extreme value approaches

A drawback with regression analysis is that, due to the inherent averaging, the fitted model is often a poor predictor of extreme values (NRC, 1991). An alternative approach, particularly useful in the context of modeling threshold exceedances, is to use extreme value theory (e.g. Gumbel, 1958; Leadbetter et al., 1983).

Cox and Chu (1993, 1996) apply a Weibull hazard model to an ozone value Y, namely

$$P(Y > y) = \exp(-(y/\sigma)^{\lambda})$$

in which the scale parameter σ is allowed to depend on meteorological variables $M_1, \ldots, M_n$ (about 100 candidate meteorological measures were used) in the form

$$\sigma = \exp\left(\Sigma\, \beta_j M_j + \zeta T \right)$$

where T is the year. The model is fitted to daily maximum hourly average ozone values for 43 Metropolitan Statistical Areas (MSAs) during 1981-1991/1993 using the average from all available station hours. The parameters are estimated using maximum likelihood, assuming independence between days. In order to take account of the daily dependence not included in the parameter estimation, standard errors are computed using a block bootstrap approach with 3-day blocks, yielding standard errors 30-40% larger than those suggested by standard likelihood asymptotics.

Niu (1996) extends the Cox and Chu approach by explicit modeling of the dependence in the errors using a heteroscedastic ARMA model with innovations scale parameter σ having the structure given in the previous paragraph, and using a nonlinear additive model for the mean field, which is also dependent on atmospheric variables. This model, while using a large number of parameters, improves on the Cox and Chu results as far as prediction of percentiles is concerned.

Joe et al. (1996), as part of the Lower Fraser Valley (British Columbia, Canada) ozone monitoring project, look for trends in high quantiles of ozone at two sites by examining days with maximum daily temperature above and below 20°C. The character of the trends are similar in both temperature ranges, although the values are, of course, larger in the higher temperature range. In order to test for trend, Joe et al. use isotonic regression (Barlow et al., 1972), i.e., testing the null hypothesis of equal means (or population quantiles) against a hypothesis of monotonically increasing (or decreasing) levels across years. For estimation of population quantiles (here the 80th and 85th) they assume lognormality and apply standard parametric likelihood methods. Dependence is modeled using an AR(1)-process.

Smith and Huang (1993) develop a logit model for threshold exceedance, where the probability p_i of exceeding the threshold on day i is given by

$$\log (p_i / 1 - p_i) = \Sigma \beta_j M_{ij}$$

The data consist of a sequence of ones and zeros, corresponding to exceedance or non-exceedance, respectively, and the likelihood is computed assuming independence between days. In fitting the model to one of the most extreme Chicago sites, using the same data as Bloomfield et al. (1993a, 1996), they find significant coefficients for year, seasonal effect, temperature, specific humidity, wind speed, and temperature–wind speed interaction. The fit was improved by introducing an indicator for whether the previous day was an exceedance, in effect turning the model into a first order Markov chain. Further lags are not needed.

Smith and Huang are also able to model the amount of excess over the threshold, using a generalization of Pickands' (1975) generalized Pareto distribution (GPD), which occurs as the limiting distribution for high level exceedance distributions under very general circumstances. Using the framework of Davison and Smith (1990) they look at models in which the logarithm of the scale parameter in the GPD is linearly related to the meteorological covariates, in a vein similar to Cox and Chu (1993, 1996) above. The results indicate significant year effect, temperature, wind speed, and temperature–wind speed interaction, much as in the analysis of exceedances. Smith (1989) considered an extreme value analysis of ozone without including covariates, and found the model to have exponential tails. In contrast, the inclusion of covariates results in a model with shorter than exponential tails. Thus, the inclusion of covariates is important for the choice of exceedance distribution.

One advantage with the extreme value approach is that one can easily compute a forecast probability of exceedance for a day, given its meteorology. This can then also be extended to compute an index for “bad ozone years,” by summing the exceedance probabilities over the days in the year to get an expected number of exceedances, based only on meteorology. Such an index can be used to assist in deciding what ozone standard violations are amenable to air pollution control strategies.

3.3 Time series filtering

Rao, Zurbenko, and colleagues have published a series of papers in recent years on meteorological adjustment of ozone data for the assessment of ozone trends and management programs (Rao et al., 1992; Rao et al., 1995; Flaum et al., 1996; Porter et al., 1996; Eskridge et al., 1997; Rao et al., 1997; Milanchus et al., 1998). Rao et al. (1997) state that statistical methods used by a number of other researchers perform poorly in situations when the changes in ozone due to meteorological variations are larger in magnitude than those induced by emissions.

The Rao-Zurbenko approach aims to separate ozone time series into three components: a synoptic-scale component attributable to weather and short-term fluctuations in precursor emissions, a seasonal scale component reflecting variation in the solar angle, and a long-term component manifesting effects of changes in climate, policy, and/or economics. We focus on two of the most recent publications of the Rao-Zurbenko methodology: Rao et al. (1997) and Milanchus et al. (1998). They consider the model

$$X(t) = b(t) + W(t)$$

where *b(t)* is a baseline component, consisting of the sum of a long-term (trend) and a seasonal variation component while *W(t)* is short-term (weather) variation. The authors analyze log-transformed ozone, so that the model accounts for multiplicative effects of weather on the baseline. The same type of decomposition is applied to meteorological time series such as temperature.

The authors separate the baseline component from the short-term variation using a computationally simple iterative application of a moving average (the KZ filter). Define

$$X_t^{(i+1)} \leftarrow \frac{1}{m}\sum_{j=-k}^{k} X_{t+j}^{(i)}$$

where $X_t^{(0)} = X_t$ and $m = 2k+1$. Then the KZ filter can be written $KZ_{mp}(X_t) = X_t^{(p)}$, where p is the number of iterations. Eskridge et al. (1997) shows that this calculation approximately filters all periods of less than $m \times p^{1/2}$ days. The authors note that other methods of decomposition, in particular using wavelets, could also be used. Thus, general issues that arise in the application and interpretation of decomposition-based analyses are not necessarily specific to the KZ filter.

The authors apply this filter to both log-daily max ozone, $O_{kz}(t) = KZ_{mp}(O_t)$, and to one or more meteorological variables, beginning with maximum daily temperature, $T_{kz}(t) = KZ_{mp}(T_t)$, with $m = 29$ days and $p = 3$ iterations.

Simple linear regressions of the filtered ozone series on the filtered temperature series shows that the relationship is stronger when the filtered temperature series is lagged; for example, a lag of 16 days for Chicago. Milanchus et al.(1998) explain this time lag by the relationship between solar angle, which peaks in late June, and the maximum surface temperature which peaks in July. However, the estimated phase shift changes greatly when the model is expanded to include an additional meteorological covariate, specific humidity. For example, the optimal time lag changes from 16 days to 7 days when specific humidity is added to the model for Chicago, and from 23 days to 44 days in a model for Cliffside Park, NJ. This casts doubt on the interpretation of this parameter as a scientifically meaningful phase shift.

Both the KZ-filtered series and the resulting short-term series are regressed on pairs of similarly decomposed meteorological variables. For the KZ-filtered meteorology, the authors switch from maximum temperature to a computed estimate of solar radiation (for which no phase lag appears necessary) and specific humidity. The short-term series covariates are maximum temperature and dew point depression.

The sum $\varepsilon(t)$ of the residuals from these two regressions are assumed to reveal changes in ozone attributable to changes in emissions. They apply the KZ filter again (with m = 365 and p = 3) to the $\varepsilon(t)$ in order to look at trends in these meteorologically adjusted residuals.

To relate this approach to simple (undecomposed) linear regression, consider the consequences of decomposing ozone $O_t = O_{lt} + O_{st}$ into a long-term and a short-term component, with a corresponding decomposition $M_t = M_{lt} + M_{st}$ for meteorology, here assumed one-dimensional for simplicity. The Rao-Zurbenko approach sets down the linear model

$$O_{lt} = \alpha_0 + \alpha_1 M_{lt} + \varepsilon_{lt}$$
$$O_{st} = \beta_0 + \beta_1 M_{st} + \varepsilon_{st}$$

yielding the resulting ozone model

$$O_t = \alpha_0 + \beta_0 + \alpha_1 M_t + (\beta_1 - \alpha_1) M_{st} + \varepsilon_t$$

indicating that this approach, unless the short-term and long-term regression slopes are identical, results in a linear model which takes into account short-term meteorology in a more sophisticated fashion than a standard linear regression model. With simple linear regression the component M_{st} would be part of the error term. Linear relationships may, of course, not capture the complexity of the ozone–meteorology association.

3.4 Spatio-temporal modeling

Carroll et al. (1997) use a spatially homogeneous and temporally stationary space-time model to study ozone exposure in Texas. Their purpose is modeling for spatio-temporal prediction, not meteorological adjustment for estimation of trend. The data come from 11 stations in the Houston area, and consist of hourly measurements of ambient ozone between 1980 and 1993. At each monitoring site, temperature, wind speed and wind direction were also measured. The modeling uses a square root transformation, and involves a deterministic trend, depending on time and temperature,

$$O_t^{1/2} = f_t(M_t) + Z_t$$

where Z_t is an error term with space-time dependence structure. The relationship with temperature is a quadratic polynomial; the authors do not use wind data, since the resulting predictions of 1993 data have higher variability than predictions without the wind data. The mean function is estimated using ordinary least squares, due to computational problems in performing the appropriate generalized least squares estimation. The authors claim that because of the large size of the data set, the loss of efficiency in using ordinary least squares should not be very important. The validity of this claim is not apparent to the reviewers.

In order to predict the spatial ozone field away from the monitoring stations, Carroll et al. use a kriging technique, with a space-time covariance function of a form that is not necessarily valid (Cressie, 1997). In discussion of the paper by Carroll et al. paper, Cressie (1997) and Stein and Fang (1997) criticize the model for not incorporating a spatial trend component. The authors respond that it would be difficult to predict this part of the trend.

While this analysis is one of the very few in the literature that explicitly incorporates spatial and temporal dependence, as well as accounting for meteorology, further development of the approach is needed for more accurate representation of the spatio-temporal structure of hourly ozone data. In particular, it is important to use valid space-time covariance structures, to incorporate meteorological variables that affect the covariance structure (such as winds), and to develop computationally feasible approaches to moderately large data sets.

The potential advantage is the ability to include atmospheric science explicitly in the modeling, which may lead to improved understanding of the processes involved.

3.5 Model assessment

In any statistical modeling, model assessment is necessary. Attention to model assessment has varied in the literature reviewed here, and some issues such as adjustment for autocorrelation have frequently been overlooked. We do not intend here to discuss and critique model assessment as it has been carried out. Rather, we want to emphasize two particular issues of some statistical sophistication: variable selection, and trend estimation.

3.5.1 Variable selection

One important issue that arises in the context of model assessment is the choice of explanatory variables to include in the model. Most assessments of variables to use in a meteorological adjustment tend to be stepwise rather than a consideration of all possible subsets, the latter being computationally challenging with large numbers of variables. Stepwise selection lacks a clear global model selection criterion, and has the problem that a variable which is eliminated early may have important interactions with other variables, which are masked by variables that are later dropped from the model (Weissberg, 1985).

A different approach, developed by Raftery et al. (1997) for linear models, and employed by Clyde (1999) in the context of health effects of particulate matter air pollution, is a fully Bayesian approach in which one sets down prior probabilities of including the various variables and then computes the associated model uncertainty in terms of posterior probabilities for a large array of models. Proponents of Bayesian model averaging argue that better predictions are obtained and that standard errors of coefficient estimates and model predictions more appropriately account for model uncertainty which is largely unaccounted for in traditional regression analysis. To our knowledge, this approach has not yet been used in the present context.

3.5.2 Modeling trend

The common overall goal of the ozone modeling considered in this review may be characterized as "minimizing the unexplained variation in ozone". The approach to achieving this goal will, however, differ, depending on the purpose of the analysis. In analyses aimed at forecasting, for instance, a major predictor of ozone is ozone at one or more previous time points (e.g. Galbally et al., 1986; Feister and Balzer, 1991). Analysis aimed at trend estimation would, on the other hand, be confounded by incorporation of lagged ozone values. In consideration of ozone trend estimation, the relevant trend is an adjusted trend, i.e., one that is not accounted for by meteorology.

Two approaches to trend estimation are possible: on the one hand one may attempt to assess the magnitude of a trend whose form has been hypothesized by, e.g., chemical and mathematical modeling. Alternatively, trend estimation may be data driven, in that one estimates the trend structure observed in the data, generally non-parametrically. Much of the trend estimation reviewed here falls into neither of these categories, but simply estimates a linear trend, without any justification. It is not difficult to imagine a situation where the presence of significant long-term structure could be masked by assuming a linear form for the investigation.

Models developed to estimate trend may also do so either by including an explicit trend term in the overall model or by assessing trend in the ozone residuals, after adjustment for meteorology, seasonality and other known sources of variation. The latter approach may lead to optimistic standard error estimates for the trend as the covariability of the trend estimate with other coefficients in the model is ignored. Alternatives to assuming a functional form for testing trend are isotonic regression (Joe et al., 1996), nonparametric regression on year within a general additive model framework (see section 4.1), using Kendall's tau on the time series of meteorologically adjusted expected ozone exceedances (Stoeckenius and Hudischewskyj, 1990), or applying rank correlation to investigate monotonic time trends in the residuals (Reynolds et al., 1998).

4. Comparison of selected methods using Chicago data

4.1 An SVD analysis

Reynolds et al. (1999) use the singular value decomposition to determine univariate summaries of the dominant patterns of association between square-root transformed ozone and nonparametrically transformed surface temperature networks in the Chicago region. The ozone network is a subset of the network analyzed by Bloomfield et al. (1996), consisting of monitors with at least 80% complete records for the 1981-1991 ozone seasons. Best subsets regression (Ryan, 1997) is used to select a linear regression model of the SVD-derived ozone summary as a function of the network temperature summary, other (possibly transformed) surface and upper air measurements, and a seasonal component. The final model includes relative humidity, a seasonal effect approximating the change in solar incident radiation, and the interaction of relative humidity with the temperature summary. Trend is investigated by adding a nonparametric trend component to the meteorological adjustment model and assessing its contribution using analysis of deviance (Hastie and Tibshirani, 1990). The trend component, while being non-monotonic, does not contribute significantly to the model, even if the residual autocorrelation is ignored. The SVD analysis reveals a strong regional scale response in ozone. The ozone network summary is approximately proportional to the network average, which is the summary obtained by Bloomfield et al. (1993a,b) from principal component analysis of the larger Chicago ozone network without direct use of meteorological fields. Bloomfield et al. (1996) base their analysis on the network median.

4.2 Trend

The SVD analysis summarized in section 4.1 estimates a non-monotonic trend which was not statistically significant. Previous analyses for the region have returned differing conclusions. Bloomfield et al. (1996) estimate a negative trend, also not statistically significant. The authors remark that the fairly wide confidence interval on the trend coefficient implies that "even strong trends may not be detected as statistically significant results for some years." Niu (1996), using similar data, obtains a significant negative trend.

Milanchus et al. (1998) estimate a non-significant positive trend at a single Chicago location. The discrepancy with the trend estimates based on regional summaries points to the difficulties with describing regional behavior using a single site. In fact, application of the Rao-Zurbenko approach to the network summary obtained in the SVD analysis results in a trend estimate comparable to that of Reynolds et al. (1999). Interestingly, a simple linear regression of ozone on meteorology applied to the single site yielded residuals with very similar structure and variability to that of Milanchus et al. (1998). In this instance there does not appear to be any advantage to applying the decomposition approach, although the implicit assumption that short-term meteorology is uncorrelated with long-term ozone (and vice versa) seems justified in the Chicago data.

Smith and Huang (1993), modeling probability of exceedance of 120 ppb at three Chicago stations, estimate negative linear trends, two of which are statistically significant. Using a threshold of 100 ppb, all three stations display a negative linear trend, but only one is statistically significant. Further analysis of this station reveals a significant negative quadratic trend in exceedances (at 120 ppb), a result that also holds for the network maximum. In both cases, a slight increase in ozone occurs up to 1984 or 1985, followed by a decrease.

The results of Smith and Huang (1993) are different from the network summary trend analyses because they are considering trends in extreme events rather than in the average ozone baseline. While shifts in average level are sufficient to produce shifts in exceedance frequency, they are not necessary for such shifts to occur. The lack of significant trends when a lower threshold was used to define exceedance is consistent with the lack of significant trends in the average ozone level.

Huang and Smith (1999) demonstrate that the strength of temporal trends can differ across meteorological conditions. While trends were predominantly negative, they were increasingly so in meteorological conditions conducive to high ozone events.

In summary, while there is general agreement that ozone in Chicago has tended to decrease over the period considered (with the exception of the positive trend found at a single site by Milanchus et al., 1998), there is considerable variability in the way trends are modeled, as well as in assessing the statistical significance of the trend estimates.

5. Discussion

Our philosophy, which has guided this assessment of the literature, is that ideally the statistical methodology should be process-driven, by which we mean that it should be guided by an understanding of the underlying physical mechanisms.

Each of the methods compared in section 4 has useful contributions to meteorological adjustment of ozone. Bloomfield et al. (1996) attempt to incorporate scientific understanding of the associations between ozone and meteorology in their nonlinear regression model. SVD (Reynolds et al., 1999) provides a sensible way of forming regional summaries for ozone and meteorology. The Rao-Zurbenko approach (Milanchus et al., 1998) allows for different associations between ozone and meteorology on different time scales. Huang and Smith (1999) distinguish trends under different meteorological conditions. Extreme value techniques (Smith and Huang, 1993; Niu, 1996), in our view, are the right approach to assess standards violations. Effects based on an assessment of mean levels are not as sensitive to tail behavior in the data, as is evidenced in the Chicago analyses.

With regard to trend estimation, the question arises as to whether a reasonable parametric form for trend can be gleaned from physical considerations. If so, statistical methods for trend estimation are well established. On the other hand, recent techniques of nonparametric function estimation, such as generalized additive models (Hastie and Tibshirani, 1990) or wavelet approaches (Brillinger, 1994), have a potential to produce not only trend estimates (which could be linear if the data warrant it), but also simultaneous confidence bands for these estimates. Such confidence bands can be used to test particular parametric models.

Summarizing non-linear trend beyond a graphical display is complicated, as it cannot be captured by single measures, such as ‘percent per decade.’ Yet, some form of summary measure is necessary to describe the trend to the public. Time-dependent summaries may be needed over the regions where non-linear trends are monotone.

It is clear that there are disadvantages to modeling regional summaries and that space-time models of the association between ozone and meteorology would be preferable. An important research area would be the development of such models for ozone extreme values.

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TABLE 1: Summary of ozone measures

TIME SCALE

5 minutely	Abdul-Wahab et al., Poissant et al., van Ooy and Carroll
Daily 30min max	Spichtinger et al.
Hourly average	Carroll et al., Cox and Chu, Fiore et al., Galbally et al., Katsoulis
Daily average	Feister and Balzer
Daily 1hr max	Bloomfield et al., Burrows et al., Davis et al., Eder et al., Flaum et al., Gao et al., Huang and Smith, Milanchus et al., Joe et al., Korsog and Wolff, McKendry, Niu, Porter et al., Pryor et al., Rao et al., Reynolds et al., Smith and Huang, Stoeckenius and Hudischewskyj, Xu et al.
Daily 8hr max	Porter et al.

LENGTH OF RECORD

Single year	Abdul-Wahab et al., Poissant et al., van Ooy and Carroll
Multiple years	Bloomfield et al., Burrows et al., Carroll et al., Cox and Chu, Davis et al., Eder et al.,, Feister and Balzer, Fiore et al., Flaum et al., Galbally et al., Gao et al., Huang and Smith, Joe et al., Katsoulis, Korsog and Wolff, McKendry, Niu, Porter et al., Pryor et al., Rao and Zurbenko, Reynolds et al., Smith and Huang, Spichtinger et al., Stoeckenius and Hudischewskyj, Xu et al.

SITES

Single site	Abdul-Wahab et al., McKendry, Poissant et al., Pryor et al.
Multiple sites, modeled separately	Burrows et al., Cox and Chu, Feister and Balzer, Fiore et al., Flaum et al., Galbally et al., Joe et al., Katsoulis, Korsog and Wolff, Milanchus et al., Rao and Zurbenko, Smith and Huang, Spichtinger et al., van Ooy and Carroll, Xu et al.
Multiple sites, univariate summary	Bloomfield et al., Davis et al., Eder et al., Gao et al., Huang and Smith, Niu, Reynolds et al., Smith and Huang, Stoeckenius and Hudischewskyj
Multiple sites, modeled jointly	Carroll et al., Porter et al.

TRANSFORMATION

None	Burrows et al., Davis et al., Feister and Balzer, Fiore et al., Gao et al., Joe et al., Katsoulis, McKendry, Niu, Poissant et al., Pryor et al., Reynolds et al. (1998), Spichtinger et al., van Ooy and Carroll
Logarithm	Abdul-Wahab et al., Bloomfield et al., Flaum et al., Korsog and Wolff, Porter et al., Rao and Zurbenko, Xu et al.
Square root	Carroll et al., Reynolds et al. (1999)
Distribution	Cox and Chu, Galbally et al.

TABLE 2: Available and included meteorology

	Chicago Analyses							Regression								
Met. variable	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Surface temp.	I	I	I	I	I	I	I	I	I	I	I	I	I	I		I
Wind speed	I	I	I	A	I	I	I	I	A		I	I	A	I		E
Wind direction	I	I	I	A	I	I	A	I	A				A	I		E
Humidity	I	I	I	I	I	I	I	A	A			I	A	I		
Pressure	A	A	I			A	A		A			I	A		I	
Radiation	I	A		IE	I	A	I	A	I			A	A			
Upper temp.	A	A				E			A				I			I
Upper wd spd.	I	I				E							A			
Upper wd dir.	A	A				E							A			
Geopotential ht.	A	A			I	E			A				A		I	I

	Regression			CART		Extreme Value				F	S	Other		
Met. variable	17	18	19	20	21	22	23	24	25	26	27	28	29	30
Surface temp.	I	I	I	I	I	I	I	I	I	I	I	I		I
Wind speed	I	A	I	I	I	I	I		I		A	I		I
Wind direction		A	I	A	I	?	I		A		A	I		I
Humidity	I	I	I	I		I	I			I				I
Pressure		A	A	I								I	I	
Radiation	I	I	I	A	I	I	I			I		I		A
Upper temp.			A	I	A	I						I		
Upper wd spd.			I	A								I		
Upper wd dir.			I	I								I		
Geopotential ht.		A		I	I	I	A					I	I	

I - incorporated into final model, A - available but not incorporated into final model, E – meteorology estimated from deterministic models; F – Filtering; S – Spatial modeling

Analyses:

1 – Bloomfield et al. (1996), 2 - Gao et al. (1994), 3 – Huang and Smith (1999), 4 – Milanchus et al. (1998), 5 – Niu (1996), 6 – Reynolds et al. (1999), 7 – Smith and Huang (1993), 8 – Adbul-Wahab et al. (1996), 9 – Feister and Balzar (1991), 10 – Fiore et al. (1998), 11 - Galbally et al. (1986), 12 – Katsoulis (1996), 13 – Korsog and Wolff (1991), 14 – Poissant et al. (1996), 15 – Pryor et al. (1995), 16 – Reynolds et al. (1998), 17 –

Spichtinger et al. (1996), 18 – Xu et al. (1996), 19 – Davis et al. (1998), 20 – Burrows et al. (1995), 21 – Stoeckenius and Hudischewskyj (1990), 22 – Cox and Chu (1993), 23 – Cox and Chu (1996), 24 – Joe et al. (1996), 25 – Smith and Shively (1995), 26 –Rao et al. (1997), 27 – Carroll et al. (1997), 28 – Eder et al. (1994), 29 –McKendry (1994), 30 -van Ooy and Carroll (1995).

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