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# Modelling dynamics and uncertainty in assessment of quality standards for fine particulate matters[*]

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**Abstract**

In order to assess compliance with air quality standards, European regulations prescribe to monitoring the concentration of particulate matters and to control both annual and daily averages.

The measurement accuracy varies according to monitor type, temperature and pollution level, often in a complex nonlinear manner. Consequently, comparisons, threshold exceedances interpretation and compliance assessment are often difficult.

In this paper, we consider the displaced dynamical calibration (DDC) model which is able to calibrate biased readings by using displaced data obtained by reference instruments. Moreover, we discuss the uncertainty of annual averages of daily concentrations. An application to the Northern Italy air quality network allows us to draw some empirical conclusions.

## 1. Introduction

The problem of setting and assessing air quality standards has a relatively long history and is related to health, epidemiological, legal, economical and social motivations.

For example in Europe, this focus is a consequence of the Treaty Establishing the European Community, Title XIX, in general and, specifically, of Council Directives 96/62/EC, on ambient air quality, and 99/30/EC, on limit values of air pollutants, including particulate matters with a diameter inferior to 10 microns ($PM_{10}$). These Directives are being gradually adopted by member states, for example in Italy this happened in the year 2002 (DM n. 02/60), and state both the air quality standards on the daily and yearly scale and the measurement quality standards for the instruments measuring $PM_{10}$.

Although it is well recognized that for health protection smaller particles ($PM_{2.5}$) are very important, monitoring practices for these particles are not yet well established in the EU, as only a "method of temporary reference for the measurement" (EC Decision, 16-jan-03) is available and the related monitoring network is poorly developed for example in the Italian territory.

In this paper, we will focus on $PM_{10}$ and consider the annual limit values for the protection of human health. These are 40 $\mu$g/m$^3$ for the annual mean and 50 $\mu$g/m$^3$ for the daily mean, not to be exceeded more than 35 times a calendar year. There are margins of tolerance (20% for the former and 50% for the latter) starting from year 2000 and decreasing linearly to 0% by January 1st, 2005.

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It is worth observing that uncertainty is explicitly mentioned in these regulations only with respect to measurement errors, precision and accuracy. For example, statistical details are implied in defining the "Reference method for the sampling and measurement of $PM_{10}$" and in demonstrating equivalence to the reference method which is based on the collection on a filter of the $PM_{10}$ fraction of ambient particulate matter and the gravimetric mass determination which we will acronymize LVG for "Low Volume sampler Gravimetric" method and/or instrument.

Contrasting with this, in this paper, while making the exercise of assessing compliance with air quality standards in Northern Italy, we will show that relevant statistical issues arise.

The first statistical issue of this paper is related to uncertainty on "annual averages" of daily particulate matters. As a matter of fact, we can see that daily $PM_{10}$ concentrations are highly auto-correlated and exhibit an empirical autocorrelation function, which is compatible with seasonal long range dependence (LRD). This is not new in high frequency air quality data, for example in ozone hourly data modelling, a seasonally fractionally integrated model has been considered by Fassò and Negri (2002a and b) and, although in depth LRD modelling of $PM_{10}$ is out of the scope of this paper, this problem will be taken into account to some extent in computing uncertainties for annual averages. Moreover, meteorological normalization of daily particulate data is important. In principle, this can be done in various ways, for example Libiseller and Grimvall (2003) discuss some problems related to meteorological normalization in the case of tropospheric ozone.

The second and main statistical issue of this paper is related to instrumental heterogeneity and space-time modelling. From our perspective, this problem is two-folded. On one side, we have short term space-time statistical modelling for particulate matters concentration, on the other side, we have the problem of calibrating possibly biased particulate readings using rare reference instruments.

Short term space-time statistical modelling for $PM_{10}$ has been recently considered by Shaddick and Wakefield (2002) and by Sun et al. (2000) from the hierarchical Bayesian point of view and, for $PM_{2.5}$, by Kolenikov et al. (2002) and Smith et al. (2003) using a nonparametric approach.

Moreover, calibration, which has recently been considered by McBride and Clyde (2003) from the Bayesian point of view for $PM_{2.5}$, arises from Northern-Italian data because of the heterogeneity of the network. As a matter of fact, in some areas, we have relatively dense networks which are based on the well known automatic monitors based on tapered element oscillating microbalance (TEOM). These monitors are known to underestimate the "true" level given by the reference method. A correction factor of 1.3 has been proposed, for example, by the APEG Report (1999).

Since, in the same area we have "some" LVG monitors, the idea of this paper is to use the LVG data to perform a dynamical calibration of the spatially displaced TEOM monitors. To do this, we will propose a state space model which can be estimated without co-located LVG data. This is of interest because TEOM has many advantages consequent to automatic operations and the capability of giving hourly data (see e.g. APEG Report, 1999).

The paper is organized as follows. In Section 2, we describe two datasets, the first is used for model validation and assessment and the second is used for discussing some problems related to practical implementation. In Section 3, we present both the co-located calibration model, which is an extension of the standard calibration model for autocorrelated data, and the displaced dynamical calibration model which is based on the Kalman filter. Section 4 introduces the problem of assessing air quality standards and discusses some problems related to the uncertainty of annual averages based on daily concentrations. The results for Piemonte and the Milan area are presented and discussed in Section 5 and 6, respectively. Section 7 concludes the paper with some comments and open problems.

## 2. The $PM_{10}$ data

In this talk we consider daily data for the years 2002 and 2003 from two important urban and surrounding areas in Northern Italy, namely Turin and Milan. The data are collected by the local environmental agencies, namely ARPA Piemonte and ARPA Lombardia.

The Milan network is characterized by six TEOM stations in/around the city of Milan in a range of $22 \times 48$ km$^2$ and the Messina Station, which is located in Milan city and is equipped with a LVG monitor. This is the typical situation where the displaced dynamical calibration model may be used.

We have some pollutants co-located in the six stations which can be used as covariates. In particular, we will use nitrogen oxides (NO) because it was found (Fassò and Nicolis, 2004) that, at least for this pollution network, it has high correlation with $PM_{10}$. Unfortunately, we do not have co-located meteorological covariates so we will use partially displaced temperatures from the Juvara and Trezzo stations.

The Turin dataset is composed of thirteen LVG monitors. In particular, three of them (Rivoli, Grassi and Consolata) are located in Turin city and one of these, Consolata Station, is also equipped with a companion TEOM gauge; another two monitors are located in the surrounding hilly area (Borgaro and Gaidano) and the remaining eight stations are spread all around the Piemonte region in an area of about $86 \times 170$ km$^2$ with most stations on the plain of the Po River and some in the neighbouring Alpine valleys (e.g. Borgosesia and Verbania) which are subject to local climatic factors.

Unfortunately, we have few covariates from the Consolata station. We will then use the co-located nitrogen oxides and Temperature from the CSELT station in Turin city. This second dataset will be used essentially for validating and testing the displaced dynamical calibration model.

## 3. Air quality calibration

Statistical calibration is broadly used in chemistry, in toxicological or immunological assays and is potentially very useful in several biometric applications (general references are in Brown 1993; Osborne, 1991; Sundberg 1999). In the standard calibration problem we have two different types of characteristics, $Y$ and $X$. The first one, $X$, is laborious or expensive to measure and, sometimes, impossible while the other is quicker or cheaper to measure but is less accurate. The statistical calibration use $X$ as an indirect measure of $Y$, that is, through the estimation of a regression model, it determines the correspondent $Y$ when $X$ has been measured.

### 3.1. The co-located calibration model

In this subsection, we consider the calibration model used as a reference for assessing the model to be defined in the next subsection.

Let $y_{t,G}$ denote the $PM_{10}$ concentration measured by the LVG monitor in $\mu$g/m$^3$ on day $t$ at the station $s$, and let $y_{t,T}$ denote the $t$-th $PM_{10}$ measurement of the TEOM monitor at the same station. In order to study the relation between these two measurements, we extend the standard linear calibration model for independent observation (see Osborne, 1991) to a time series with autocorrelated errors.

We then have

$$y_{t,G} = \alpha + \beta y_{t,T} + \theta' x_t + e_t \qquad (1)$$

where $\alpha$ and $\beta$ represent, respectively, the additive and multiplicative relative bias for the TEOM measurements on day $t$, $\theta$ *is* the regression coefficient vector and $x_t$ is a vector of covariates including, in our case, daily averages of temperature and nitrogen oxides concentration. We then assume that the error $e_t$ is a Gaussian ARMA process.

After estimating the parameter on a calibration sample ($x_t, y_{t,G}, y_{t,T}$), using this model, the gravimetric $PM_{10}$ values may be estimated by the available TEOM and covariates.

### 3.2. The displaced dynamical calibration model

In this section, we consider the case of practical interest in which $y_{t,G}$ and $y_{t,T}$ are gathered at two displaced stations, *s* and *s*' say. The Displaced Dynamical Calibration model (DDC) for the $PM_{10}$ concentrations is expressed as a state space model (see Harvey, 1989; Durbin and Koopman, 2001; Brown et al., 2001) with the following measurement equations

$$\begin{aligned} y_{t,G} &= \mu_t + \varepsilon_{t,G} \\ y_{t,T} &= \alpha + \beta(\mu_t + \delta_t) + \varepsilon_{t,T} \end{aligned} \qquad (2)$$

and state equations

$$\begin{aligned} \mu_t &= \Phi\mu_{t-1} + \eta_{t,\mu} \\ \delta_t &= \omega + \theta' x_t + \eta_{t,\delta} \end{aligned} \qquad (3)$$

for $t = 1,\ldots, T$. The quantity $\mu_t$ is the unobserved "true" mean $PM_{10}$ concentration on day $t$ at station *s* and depends on the past? through the coefficient $\Phi$. Similarly, the quantity $\mu_t+\delta_t$ is the corresponding "true" concentration for station *s'*. Hence $\delta_t$, being related to the "local" covariates $x_t$ as above, describes the local features of station *s'*. When $\theta$ is zero we have $E\delta=0$, and $\delta$ may be interpreted as a random effect; in our case $E\delta=\theta' x$ may be interpreted as the local differential?level. Moreover, $\alpha$ and $\beta$ have the same calibration interpretation as equation 1. Finally, we assume the usual normality distribution for the measurement errors.

It follows that the unobserved component

$$\hat{y}_t = \mu_t + \delta_t \, ,$$

which may be computed using the Kalman filter, represents the "calibrated" TEOM measurements of $PM_{10}$ concentration in station *s'* and may be computed using displaced LVG data together with co-located TEOM and covariates.

In many applications, log-transformed data are used in order to fit such data. In this application we found that, although PM10 data generally have skew distribution, probably due to the error measure nature of this model, the residuals of both measure and state equation 2 and 3 above show acceptably normal distribution. Moreover, they do not show autocorrelation nor heteroskedasticity.

## 4. Annual average uncertainty and standards

When computing annual averages it raises the issue of uncertainty and standard deviation computation. If we consider daily $PM_{10}$ data as a purely random time series we are led to consider the "total variance" of the annual average $\bar{y}$. Alternatively, we may be interested in assessing the uncertainty of $\bar{y}$ keeping the seasonal effect fixed. In both cases, we have to take into account the effect of autocorrelation. To do this, we use the variance formula for the sample average of stationary correlated data:

$$Var(\bar{y}) = \frac{Var(y_t)}{n} + 2\sum_{j=1}^{n-1} \frac{(n-j)}{n^2}\gamma(j) \tag{4}$$

where $\bar{y}$ is the annual average ($n$ = 365) and γ(j) are the autocovariances which can be estimated on the full data set (in our case $N$ = 730).

By computing the total variance in this way, we get rather large std($\bar{y}$) between 4 and 10 $\mu g/m^3$. To improve this, the standard deviations reported in the tables of the following sections are based on adjusted data. In our case, since we have just two years of data and only limited data on meteorology, we try to adjust the $PM_{10}$ with respect to observed daily temperatures which may be taken as a proxy of seasonal and other meteorological factors.

We then use the regression model with correlated errors previously introduced with the aim of calibration in equation 1. Here, of course, it is used with generic $y_t$ term and without the calibrating component ($\beta = 0$). The variance formula (4) has been applied to the corresponding correlated residuals giving, say, $Var(\bar{e})$. Using **S** for seasonality and meteorology, we than get the approximated relation

$$Var(\bar{y}|\mathbf{S}) = Var(\bar{e}).$$

Moreover, when $y_t$ is the output of a calibration model this quantity is based on the calibration error variance which, in the DDC case, is an output of the Kalman filter. Finally, uncertainty on model parameter estimation should be added but this last point has been omitted for simplicity here and is deferred to a future paper.

## 5. Fitting and assessing the DDC model

In this section, using the LVG network with a co-located TEOM monitor from ARPA Piemonte, we discuss the reconstructing capability of the dynamical calibration model in comparison to the "1.3 rule" and the reference calibration regression equation 1 with ARMA(1,1) errors and covariates given by temperature and nitrogen oxides.

In particular, we fit a DDC model for each station with TEOM data from Consolata. The estimated parameters, given in table 1, show that the dynamic parameter Φ is essentially constant over space. This is consistent with the findings of Shaddick and Wakefield (2002) based on hierarchical modelling. Differently, but not surprisingly, the intercept α has a marked variation between stations. We note that α is strictly related to the station average level.

Examining the Turin-city-stations (Borgaro, Grassi, Rivoli and Gaidano) we see more homogenous behaviour, with all but Grassi having similar β coefficients.

Moreover, using LVG data from Consolata, we are able to assess the model performance. This is done by comparing the behaviour of the "reconstructed data" $\hat{y}_t$ and the observed $y_t$, that is the LVG data taken as the "true $PM_{10}$ value" and ignoring the error component of equation 2. The corresponding $R^2$ are reported in table 2 for DDC models with and without covariates (DDC-X). We see that local temperature and nitrogen oxides together improve the fitting substantially. This is especially true when the displaced LVG calibrator is far away (see Verbania and Borgosesia).

Only the Grassi Station show better fitting without covariates and this may be due to the fact that different stations have equal or different local levels. We found that working with the mean deviations still improves fitting and eliminates this anomaly but this approach has not been studied in depth here because it requires an estimate of the LVG mean at the calibrating station *s'*.

**Table 1**. Estimated DDC models in Turin and Piemonte

| | $\alpha$ | Std($\alpha$) | $\beta$ | Std($\beta$) | $\Phi$ | Std($\Phi$) | $\theta_1$ (NO) | Std($\theta_1$) | $\theta_2$ (Temp) | Std($\theta_2$) |
|---|---|---|---|---|---|---|---|---|---|---|
| Alba | 32.49 | 2.94 | 0.46 | 0.03 | 0.97 | 0.01 | 0.06 | 0.04 | 0.27 | 0.26 |
| **TO Borgaro** | 22.46 | 2.42 | 0.58 | 0.03 | 0.97 | 0.01 | 0.11 | 0.03 | 0.51 | 0.17 |
| Borgosesia | 28.41 | 3.42 | 0.51 | 0.04 | 0.97 | 0.01 | 0.18 | 0.04 | 0.43 | 0.25 |
| Bra | 24.57 | 2.77 | 0.48 | 0.03 | 0.97 | 0.01 | 0.06 | 0.04 | 0.30 | 0.22 |
| CN II Regg. Alpini | 39.79 | 3.15 | 0.35 | 0.04 | 0.96 | 0.02 | 0.26 | 0.06 | -0.70 | 0.39 |
| Saliceto | 41.22 | 4.08 | 0.27 | 0.05 | 0.97 | 0.01 | 0.40 | 0.11 | -0.59 | 0.69 |
| **TO Grassi** | 21.46 | 1.16 | 0.47 | 0.01 | 0.96 | 0.01 | | | | |
| **TO Rivoli** | 17.67 | 2.10 | 0.56 | 0.02 | 0.98 | 0.01 | 0.05 | 0.02 | 0.94 | 0.14 |
| **TO Gaidano** | 23.36 | 2.49 | 0.57 | 0.03 | 0.97 | 0.01 | 0.01 | 0.03 | 0.90 | 0.17 |
| VC Gastaldi | 41.41 | 3.80 | 0.31 | 0.04 | 0.98 | 0.02 | 0.10 | 0.08 | -1.14 | 0.51 |
| Verbania | 33.88 | 3.52 | 0.44 | 0.04 | 0.96 | 0.01 | 0.29 | 0.05 | -0.29 | 0.35 |
| NO Leonardi | 29.16 | 2.63 | 0.38 | 0.02 | 0.95 | 0.01 | 0.25 | 0.05 | -0.12 | 0.30 |

**Table 2.** Comparison of DDC models with and without covariates in Turin and Piemonte.

| | 100x$R^2$ | | |
|---|---|---|---|
| | DDC | DDC-X | good data |
| Alba | 41.3% | 63.7% | 71.9% |
| **TO Borgaro** | 49.8% | **82.3%** | 74.4% |
| Borgosesia | 6.5% | 70.4% | 58.2% |
| Bra | 69.8% | 78.0% | 68.8% |
| CN II Regg. Alpini | 15.4% | 30.9% | 77.0% |
| Saliceto | 18.2% | 25.5% | 65.1% |
| **TO Grassi** | **84.2%** | 43.0% | 75.5% |
| **TO Rivoli** | 72.9% | **82.8%** | 61.4% |
| **TO Gaidano** | 45.0% | **78.9%** | 63.7% |
| VC Gastaldi | 64.1% | 31.2% | 54.9% |
| Verbania | 5.3% | 58.9% | 65.8% |
| NO Leonardi | 72.4% | 75.9% | 76.0% |

Using the year 2003 as a reference, in table 3, we consider in more detail the behaviour of DDC in Turin city. In particular, the number of exceedances of the the annual limit value for the protection of human health together with the percentages of correct-over-effective and false-over-predicted exceedances are compared for some alternative models. In the first line, we find the "true" data, in the second line the performance of the "1.3 rule" discussed in the introduction, and in the third line the co-located calibrator from equation 1.

We see that the fitting performance ($R^2$) of the DDC model is between the "1.3 rule" and the co-located calibration model. Moreover, the annual averages and exceedances are less extreme than "1.3 rule".

Finally, we note that the standard deviations of the annual average of DDC and co-located calibration data are smaller than observed TEOM and LVG. This is due to the use of error variance from calibration model which filter out not only the seasonal component, but also components at any frequency and the TEOM component. We recognize that using different uncertainty assessments in the same table may not be the best practice for direct comparisons. Nevertheless, this gives, in some sense, an idea of the extent of the omitted variance component due to model estimation.

**Table 3.** Summary of the reconstructing capabilities in Turin, Consolata, year 2003.

| | | | | Exceedances | | | |
|---|---|---|---|---|---|---|---|
| | Mean | std | Bias | Detected | Correct | False | $R^2$ |
| LVG Consolata | 63.6 | 2.10 | | 166 | | | |
| 1.3 rule | 73.2 | 1.30 | 9.6 | 265 | 96.9% | 38.0% | 75.2% |
| Co-located calibration | 65.6 | 0.62 | 2.0 | 187 | 90.3% | 13.2% | 88.8% |
| *DDC Models:* | | | | | | | |
| Borgaro | 55.39 | 0.99 | -8.2 | 134 | 76.6% | 5.0% | 82.3% |
| Grassi | 70.7 | 1.45 | 7.1 | 193 | 94.4% | 19.6% | 84.2% |
| Rivoli | 61.86 | 0.89 | -1.7 | 215 | 92.0% | 30.8% | 82.8% |
| Gaidano | 51.52 | 0.82 | -12.1 | 115 | 67.5% | 6.7% | 78.9% |

## 6. Data analysis of Milan area

In this section, we apply the DDC model to Milan data in order to assess air quality standard attainment. In table 4, the reported estimated parameters have interpretations paralleling table 1. Differently from Consolata results, here nitrogen oxide is not useful and we use only temperature. Although the corresponding *p-values* are not very small, we keep all covariates in this case in order to "surrogate" the local mean level which, once again, is not available.

In table 5, where standard deviations are computed similarly to table 3, the annual averages and exceedances are reported respectively for the raw TEOM data, the "1.3 rule" and the DDC models of table 4. Moreover, in the last line, the data for the "true" concentrations of Messina station are reported.

The DDC model gives annual averages which are close to "1.3 rule" in the city center (line labels prefixed with MI-) and lower values in the surrounding areas. Moreover, the number of exceedances given by DDC is always less high than "1.3 rule".

**Table 4**. Estimated DDC models in Milan.

| | $\alpha$ | Std($\alpha$) | $\beta$ | Std($\beta$) | $\Phi$ | Std($\Phi$) | $\theta_1$ (temp) | Std($\theta_1$) |
|---|---|---|---|---|---|---|---|---|
| Magenta | 17.61 | 2.13 | 0.44 | 0.02 | 0.94 | 0.02 | -0.33 | 0.20 |
| MI Juvara | 13.38 | 1.80 | 0.53 | 0.02 | 0.95 | 0.02 | -0.12 | 0.13 |
| MI-Verziere | 12.60 | 1.73 | 0.50 | 0.02 | 0.96 | 0.01 | 0.09 | 0.13 |
| Pioltello | 17.73 | 1.94 | 0.43 | 0.02 | 0.96 | 0.01 | -0.61 | 0.19 |
| Meda | 25.70 | 2.49 | 0.37 | 0.03 | 0.96 | 0.01 | -1.02 | 0.30 |
| Vimercate | 15.51 | 1.89 | 0.41 | 0.02 | 0.95 | 0.02 | -0.40 | 0.19 |

**Table 5**. Annual averages and exceedances in Milan, year 2003.

| | Means | | | | | | Exceedances | | |
|---|---|---|---|---|---|---|---|---|---|
| | TEOM | Std.Dev. | 1.3 rule | Std.Dev. | DDC | Std.Dev. | TEOM | 1.3 rule | DDC |
| Magenta | 47.20 | 2.03 | 61.35 | 2.64 | 56.50 | 1.49 | 93 | 171 | 137 |
| MI-Juvara | 47.62 | 1.96 | 61.91 | 2.54 | 58.76 | 1.26 | 101 | 169 | 138 |
| MI-Verziere | 46.08 | 2.62 | 59.90 | 3.40 | 61.99 | 1.22 | 85 | 166 | 155 |
| Pioltello | 45.12 | 2.22 | 58.65 | 2.88 | 53.80 | 1.33 | 88 | 147 | 130 |
| Meda | 47.77 | 1.58 | 62.10 | 2.05 | 48.48 | 1.56 | 89 | 175 | 118 |
| Vimercate | 42.17 | 1.74 | 54.83 | 2.26 | 55.14 | 1.29 | 64 | 137 | 129 |
| *LVG* | | | | | | | | | |
| MI-Messina | 61.98 | 2.29 | | | | | 147 | | |

## 7. Conclusions and further developments

We have discussed two statistical issues arising in assessing the attainment of air quality standards. In particular, we have considered some techniques for assessing uncertainty of annual averages. This approach is useful also for many other pollutants and admits a number of variants in the field of seasonal modelling and decomposition. The second and main issue addressed in this paper is the correction of $PM_{10}$ readings based on TEOM monitors. We have shown that, using displaced gravimetric monitors it is possible to have good calibrated data and, at least for our data, the dynamical displaced calibration model does improve with respect to the simple "1.3 rule" and approaches the optimal co-located calibration model.

The DDC model, allow for missing values handling, covariates and dynamic adjustment. Moreover, it can be substantially improved if partial information is available about local averages, obtained, for example, by a mobile station.

Of course many open problems require further solutions. Among these, we mention two areas, the first is related to a problem which has been completely ignored in this paper, i.e. the uncertainty assessment on the number of exceedances which are fixed by European regulations at 35, five days per calendar year.

The second is related to the DDC model and calls for many further extensions, including model averaging, long range dependence modelling and space-time modelling.

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