Random and Dynamical Calibration for Air Quality Measurement Instruments

Taratura aleatoria e dinamica di strumenti di misura della qualità dell'aria

I. Negri[1], S. Salini[2]

[1] Dipartimento di Ingegneria Gestionale e dell'Informazione
Università degli Studi di Bergamo, ilia.negri@unibg.it
[2] Dipartimento di Scienze Economiche Aziendali e Statistiche
Università degli Studi di Milano, silvia.salini@unimi.it

Riassunto: In questo lavoro si cerca di ricavare il valore della concentrazione nell'aria delle polveri sottili, dal diametro minore di 2.5 micron (PM2.5), da quello delle polveri di diametro fino a 10 micron (PM10). Il monitoraggio di quest'ultima è effettuato ormai da anni in tutti i paesi occidentali e costituisce uno dei parametri per la valutazione della qualità dell'aria. Ciò non accade invece per le PM2.5, che risultano anche più pericolose per la salute dei soggetti più deboli e più a rischio di malattie cardio-polmonari. Tuttavia, dal momento che la concentrazione delle PM2.5 riguarda una percentuale che varia dal 60% al 90% delle PM10, il problema può essere affrontato con i modelli di taratura stocastica dinamica in cui il valore delle PM2.5 è considerata la misura incognita, da ricavare partendo da un campione di taratura.

Keywords: Fine and Coarse Particulate Matters, Kalman Filter, Statistical Calibration.

1. Introduction

The classical calibration problem of a measurement instrument is approached, from a statistical point of view, with linear models strongly dependent on the so called, calibration sample. These kinds of models fit very well static calibration situations, such that encountered in an engineering context, where the explicative variables are not random variables and the calibration problem of a measurement instrument is based on a preliminary design of the experiment.

The measurement of the airborne particulate matter (PM) presents many difficulties. First of all PMs originate from a variety of sources and have morphological, physical, thermodynamic and chemical different properties that are very difficult to capture with a single measure instrument. Moreover the composition depends also on temperature, pressure, relative humidity and other climatic conditions that may influence the measure. As a consequence difficulties arise in determining the precision and the accuracy of a PM monitoring technique. In particular no standard reference calibration material or procedure has been developed for suspended PM. This work, following the theory of statistical calibration, is a first attempt trying evaluete the PM2.5 concentration (PM with a diameter less than 2.5 micron) as a function of the PM10 (PM with a diameter less than 10 micron). The interest in PM2.5 is related to the fact that those matters are the most dangerous for human health and it is under discussion their

inclusion in the air quality standard figures for particulate matter in the UE. Therefore PM2.5 concentration need to be monitored and our proposal goes in this direction. Unfortunately in a complex and mutable context such that described above, the classical statistical calibration approach appears inadequate and models based on random and dynamical calibration seem to be more reasonable. We propose a dynamical approach based on Kalman filter (Salini et al., 2002) trying to improve the performance of the classical static linear estimator. Dynamical calibration models have been recently applied in the context of quality standards for particulate matters (Fassò and Nicolis, 2004) in a state-space framework.
The next section concerns the airbone particulate matter generation, its dangerous effects on human health and the air quality national standards for these air pollutants.
Section three presents the random calibration model and the dynamical calibration models.

2. Airborne particulate matter

Particulate matter is a complex mixture consisting of varying combinations of dry solid fragments, solid cores with liquid coatings and small droplets of liquid. These tiny particles vary greatly in shape, size and chemical composition, and are made up of many different materials such as metal, soot, soil and dust. Atmospheric particles contains also inorganic ions and hundreds of organic compound. PM originate from different sources. Examples include combustion generate particles, such as diesel soot or fly ash, photochemically produced particles, salt particles and soil-like particles from resuspended dust. Airborne particulate matter has both a primary component, which is emitted directly from sources such as road traffic industry, and a secondary component which is generated in the atmosphere by chemical reactions of gases, mainly sulphur dioxide, oxides of nitrogen and volatile organic compounds.
The aerosol community uses a different approach to the PM classification by size. PM with an aerodynamic diameter below 10 microns are denoted PM10. They are known also as inhalable or thoracic particles as they are able to enter the respiratory tract including the head airways, the larynx and the lung. The term fine particulate matter is reserved for the particles having an aerodynamic diameter inferior to 2.5 micron (denoted with PM2.5). They are called breathing particles and are able to reach the gas-exchange region of the lung. The coarse component of the PM is made up by the particles with diameter less than 10 micron but greater than 2.5 micron (Wilson et al. 2002).
Fine and coarse particulate matters differ not only in size but also in the generation mechanisms, sources, toxicity and health effects. Fine particles are mainly produced by combustion or burning activities such as fuel emitted by automobiles, factories, fireplaces, and wood stoves. Coarse particles usually arise as a result of natural processes such as wind-blown dust or soil. These particles may produce harmful health effects such as worsening of heart and lung diseases, being very young people as well as elderly people the most exposed to the risk. Exposure to elevated concentrations of PM is also associated with increased stays in hospital and doctor visits and increased numbers of premature deaths.
Recently the total suspended particles (TSP) measurement has been suspended in many country in favour of the PM10 monitoring (see e.g. US National Ambient Air Quality

Standard for the particulate matter in the Federal Register 1987). Consequently, the US government has adopted an air quality standard for particles measured as PM10 of 150 micrograms per cubic meter (μg/m3) expressed as a 24 hour running mean (the average of any consecutive 24 hourly measurements at an individual site) and an annual average limit of 50 μg/m3. The European Union has recently agreed with limit values for PM10 of 50 μg/m3 measured over fixed 24 hour periods, not to be exceeded more than 35 times per year (equivalent to a 90th percentile compliance with 50 μg/m3), and an annual average limit value of 40 μg/m3, both to be achieved by the year 2005. There are no air quality standard for the PM2.5 in the UE. In the US the EPA air quality standards have fixed an annual average of 15 μg/m3 and 24-hour average of 65 μg/m3. The state of California has adopted revised PM standards, by lowering the annual PM10 standard from 30 μg/m3 to 20 μg/m3 and establishing a new annual standard for PM2.5 of 12 μg/m3.

3. Random and Dynamical Calibration

Calibration is the process whereby the scale of a measure instrument is determined or adjusted on the basis of a calibration experiment. Statistical calibration is a kind of inverse prediction, broadly used in chemistry, engineering, biometrics and is potentially useful in several practical applications. A review can be found in Osborne (1991).
Suppose that two different instruments for the measurement of the same quantity are considered, the first one (*standard method*) being more laborious, accurate and expensive than the second one (*test method*). The variables associated to the measures obtained by the two instruments are indicated by X and Y respectively. A sample of n units, in which both measures x and y are observed, is considered. The set of values (x_i, y_i) i=1,...,n is called calibration experiment. The statistical calibration problem arises when only the y_i obtained by the test method are known and the unknown x_i have to be estimated. The solution of this problem depends on the hypotheses on the probabilistic model supposed to have generated the calibration experiment. In this paper it is assumed that (x_i, y_i) are realizations of a bivariate random variable (*random calibration*), whose components are linearly related, according to the following calibration model: $Y = \alpha + \beta X + \varepsilon$ where $\varepsilon \sim N(0,\sigma^2)$. The MLE classical estimator is $\hat{x}_C = (y - \hat{\alpha})/\hat{\beta}$ in which $\hat{\alpha}$ and $\hat{\beta}$ are the OLS estimators of α and β (Brown, 1993).
We consider the PM2.5 being the standard measure X and PM10 the test measure Y. As mentioned in the introduction the static classical statistical calibration approach appears inadequate in this context, since the X measures show a dynamical behaviour, usually described by the following equation: $x_{t+1} = Ax_t + B\mathbf{z}_t + \eta_t$ where η_t is an error component and $\mathbf{z}_t$ is a set of exogenous covariates.
In a previous work a dynamical calibration model based on Kalman filter is proposed (Salini et al. 2002). Kalman filter solves the problem of recursively estimate the state of a dynamical system given its measures. The classical linear estimator is the prior estimator of Kalman filter algorithm and the posterior estimator is the classical one adjusted with the Kalman gain and the Kalman innovation that depends on the covariance of measurement error (difference between true measure and test measure) and on the covariance of estimation error (difference between true measure and classical estimators). The limit of this model is that changes of the true measure over time are

not considered, while it is well known that the concentration of particulate matter is highly dependent on previous measures. So we propose a new time dependent model based on Kalman filter. Following the usual notation of calibration theory, we denote PM2.5 level at time *t* with and PM10 measure at time *t* with .

The reference model considers both dynamical equation for the PM2.5 and the relation between PM10 and PM2.5, governed by the linear model , in according with the classical calibration approach. Generalizing the Kalman filter algorithm we obtain a new estimator that differs from the classical one and from the one proposed in Salini 2002, because it depends also on covariates and it is adjusted using the previous estimated measure or, when known, the true measure.

4. Results and Discussion

The standard calibration model and the proposed dynamical calibration model are applied on the data of PM10 and PM2.5 concentration measured in a station in Rimini (Italy). The models are tested for daily measures taken on a six months period. In Table 1 are shown results.

	Errors Mean	Errors Standard Deviation
Classical Estimator	0.00	7.23
Kalman Estimator	1.13	2.40
Dynamical Estimator	0.71	2.10

Table 1: *Comparison of models. Third model considers seasonal components.*

We can note haw Kalman estimator reduces drastically the errors standard deviation but introduces a bias. We attempt that introducing seasonal components and covariates will further improve the performance of prediction. To test our proposed model we will utilize data of Harrison Park of Berkeley in California during 2002 available at http://www.ci.berkeley.ca.us/parks/parkspages/HarrisonAirQuality.html and data giving us by ARPA Emilia Romagna.

References

Brown, P. J. (1993) *Measurement, Regression and Calibration*, Oxford University Press, Oxford.

Fassò A., Nicolis O. (2004) Modelling dynamics and uncertainty in assessment of quality standards for fine particulate matters, *GRASPA Working paper* n. 21.

Janssen L.H.J.M, Buringh E., van der Meulen A., van den Hout K.D., (1999), A method to estimate the distribution of various fractions of PM10 in ambient air in the Netherlands, *Atmospheric Environment* 33, 3325-3334.

Osborne, C. (1991) Statistical calibration: a review. *International Statistical Review*, 59, 309-336.

Salini S., Zirilli A., Tiano A. (2002) Multivariate Calibration by means of Kalman filter, in *proceedings SIS 2002*, 5-7 Giugno, Cleup Editrice, pp. 493-496.

Wilson, WE et al. (2002), Monitoring of particulate matter outdoors, *Chemosphere*, 49, 1009-1043.