DISPLACED CALIBRATION OF PM₁₀ MEASUREMENTS USING SPATIO-TEMPORAL MODELS

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1. INTRODUCTION

Interest in the investigation of airborne particulate matter (PM) concentration is a key issue in environmental monitoring, mainly due to the fact that several epidemiological studies have shown a link between daily PM levels and adverse health effects (see for example Pope *et al.*, 1995). Hence the public authorities' decision to create monitoring networks designed to monitor PM levels in given administrative regions.

The said monitoring networks very often suffer from two important flaws: firstly, their location is almost always established without using statistical optimisation criteria; secondly, the monitoring sites are often equipped with heterogeneous measuring instruments. Low Volume Gravimetric (LVG) samplers represent the current benchmark instrument for the sampling and measurement of PM, according to the European Community directives adopted in Italy in 2006 (DM 02/06). For this reason, air quality standards are set according to LVG samplers. Such samplers work by collecting the PM₁₀ fraction of ambient particulate matter on a filter and then determining the gravimetric fraction. Despite the fact that LVG samplers are widely agreed to be the most effective instruments for measuring PM₁₀ concentration, the monitoring sites are very often equipped with inefficient automatic samplers such as the Tapered Element Oscillating Microbalance (TEOM) sampler. Such instruments are known to underestimate the true level given by the reference method. However, TEOM samplers have the advantage over LVG samplers in that they automatically record hourly data (whereas LVG samplers only take daily measurements). For this reason, TEOM measurements need to be "calibrated". Calibration consists in the transformation of TEOM measurements in order to obtain reliable estimates of true pollution levels. The parameters used for the purposes of this process of transformation should be estimated using data collected by co-located LVG and TEOM measuring instruments in a suitably designed experiment. In recent years, TEOM data have generally been corrected by applying a 1.3 multiplicative correction factor to the measurements (the "1.3 rule"), thus implying a linear relationship between LVG and TEOM

measurements. The 1.3 correction factor has been proposed, for example, by the APEG report (1999).

In recent years, a number of papers have been published on the subject of the spatio-temporal modelling of PM data recorded by monitoring networks (see Shaddick and Wakefield, 2002; Smith et al., 2003; Riccio et al., 2006; Cocchi et al., 2006). The space-time modelling of air pollutants is a complex process due to the fact that temporal dependence, spatial dependence and interaction between spatial and temporal behaviour, have to be modelled simultaneously, together with the dependence on meteorological covariates and other explanatory variables. Hierarchical models are an effective instrument with which to build complex models from relatively simple sub-models. The Bayesian framework is the most suitable one for managing spatio-temporal models involving complex relationships (Wikle et al., 1998). Cocchi et al. (2006) proposed a spatio-temporal Bayesian hierarchical model for daily mean concentrations of PM₁₀ measured at 11 monitoring sites located in the main cities of the Emilia-Romagna Region, over a three-year period. All the monitoring sites included in the analyses were equipped with LVG samplers. The present paper proposes that this model be extended in order to deal with data collected by heterogeneous measuring instruments located within the same administrative area. We show how the model can be employed to produce calibrated measurements for those sites equipped with TEOM samplers. One of the aims of this model is to obtain a more effective rule than the "1.3 rule", with which to calibrate TEOM measurements. We point out that an efficient calibration procedure is needed since environmental standards are set according to the LVG sampler. A further aim of the model is to assess compliance with environmental standards at sites equipped with TEOM samplers.

The classical problem of calibration (see for example Brown, 1994) involves measurements of the same quantity, along time and space, performed simultaneously by a reference and an equivalent measuring instrument. Our case is different, however, since reference and equivalent instruments are not located at the same points. Thus our approach produces "pseudo-calibrated" measurements since it is not based on co-located LVG and TEOM devices: this problem is defined in the literature as "displaced calibration" (Fassò and Nicolis, 2005). The approach is feasible when spatial correlation is taken into account: in fact, reference measurements can only be used to calibrate non-reference measurements at far-away monitoring sites if a correlation structure exists in the pollution field. In studies of air-pollution levels, this is possible because of the spatial correlation and because of the common temporal trend of the pollution field. A non-Bayesian approach based on the application of a Kalman filter to a state-space model is proposed in Fassò *et al.* (2005) for the PM₁₀ monitoring network (equipped with LVG and TEOM samplers) in the Piedmont Region.

The current paper is organised as follows: Section 2 describes the data set that led to the development of the hierarchical model; Section 3 provides an outline of a general hierarchical model designed for networks equipped with homogeneous measuring instruments; this model is extended in Section 4 in order to cope with heterogeneous monitoring networks. Finally, in Section 5, we discuss the results

of the application, the inadequacies of the "1.3 rule", and the assessment of air quality at monitoring sites equipped with TEOM samplers, according to the proposed model output.

2. THE MOTIVATING DATA SET

The analysed data set consists of time series of PM_{10} daily means ($\mu g/m^3$) collected at 12 monitoring sites located throughout the largest urban areas in the Emilia-Romagna Region, Italy; Figure 1 shows the spatial location of the said monitoring sites. The study period extends from January 1st 2000 to December 31st 2002. Although measurement of PM_{10} only started in 1998, a satisfactory spatial coverage of the region had been achieved by the beginning of the chosen study period. At least one monitoring site is available for each of the 9 administrative districts within the region, whose borders are shown in Fig. 1.

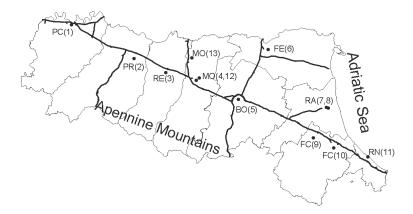


Figure 1 – The monitoring sites' spatial location. The bold lines indicate the main roads, while the dots indicate the monitoring sites locations: the letters are abbreviations for the names of the towns, while the monitoring site identifying numbers are shown in brackets.

The monitoring sites - with the exception of numbers 6, 7 and 8 - are located along the main arterial road (the Roman Via Emilia) joining the region's main towns and cities. The Apennine Mountains lie to the South of the Via Emilia, and the entire area south of that line is devoid of all significant industrial settlements. Sites 7, 8 and 11 are located in towns near, or on, the Adriatic coast. Site 12 is the one monitoring site equipped with a TEOM sampler. The other monitoring sites are equipped with LVG samplers. Site 13 is an additional monitoring site equipped with a TEOM sampler but not included in the model estimation. Data collected at this site are used to evaluate model performance in predicting off-sample PM₁₀ data.

The percentage of missing values varies from 7% to 40% across the monitoring sites. Missing values can be treated as parameters within the Bayesian framework. Such values can be inferred by integrating out model parameters from the distribution of missing data given the observed values.

The sites have been sub-divided according to their specific urban location: 4 are located in background urban areas such as parks (Type A), while the remaining 7 are located in densely populated areas or areas with high traffic density (Type B and C). Site 12 is located in a background urban area (Type A). A preliminary explorative analysis (not reported here) revealed that PM₁₀ levels are, on average, lower at Type A monitoring sites, while the levels at Type B and C sites are comparable to each other.

The time series seasonality is very similar for all three types. There is a strong linear correlation between site measurements, ranging from 0.86 for the nearest sites to 0.6 for those further away. The observed correlations remain high for the site equipped with the TEOM sampler. Even though the correlation decreases slightly as the distance increases, there is nevertheless a strong correlation between distant monitoring sites' time series measurements, indicating that a considerable amount of the between-sites correlation is due to the common temporal dynamics of data.

Meteorological variables for each site are obtained from the mass-consistent CALMET model used by the Emilia-Romagna Regional Meteorological Service. The use of predicted meteorological variables provides homogeneous covariates for each monitoring site, which would not be otherwise available if data were obtained by actual measurement, thus avoiding the problem of spatial misalignment as well as missing values in covariates. The model offers estimates of daily mean temperature, daily mean mixing height (MH) and daily mean wind speed (WS), on a regular grid of $10km \times 10km$. The underlying reasons for the choice of MH instead of temperature (which is more widely used in air pollution modelling) are discussed in some detail in Cocchi *et al.* (2006). For the purposes of model estimation, the logarithmic transformation needs to be applied to PM₁₀ data in order to obtain an approximately symmetric distribution for each monitoring site, and to stabilize the mean-variance relationship.

3. THE HIERARCHICAL MODEL FOR A HOMOGENEOUS NETWORK

Let Y_{ts} and M_{ts} denote the log-PM₁₀ concentration and the vector of meteorological covariates at spatial location s on day t (t=1,...,T) respectively, and let (C_{1s} , C_{2s}) be the geographical coordinates of site s (s=1,...,S). In what follows, first and second subscripts denote time and space dimensions, respectively. A dot as a subscript indicates that the whole domain referred to by the subscript is taken into consideration. We assume that:

$$Y_{t \cdot} \mid \mu_{t \cdot}, \sigma_{\cdot}^2 \sim MVN(\mu_{t \cdot}, diag(\sigma_{\cdot}^2))$$
 (1)

where Y_t and μ_t denote the *S*-dimensional vectors of the observed log-PM₁₀ concentration at time t, and the unknown values of concentrations at the same time, respectively. Moreover, $\sigma_{\bullet}^2 = (\sigma_1^2, \sigma_2^2, ..., \sigma_S^2)$ represents the vector of resid-

ual variances at the S monitoring sites. Conditionally on model parameters, observations at time t are independent. Unknown mean levels are modelled as follows:

$$\mu_{tc} = \gamma_1 Z_c + \gamma_2 C_{1c} + \gamma_3 C_{2c} + \mathbf{M}_{tc}' \delta + \theta_t + \varepsilon_{tc}$$
(2)

where $Z_s = 1$ if site s is located in a background urban area (Type A), and $Z_s = -1$ if site s is located in an area with high traffic and population densities (Types B and C); hence parameter γ_1 measures the effect of the monitoring site type on the average log-PM₁₀ concentration. Parameters γ_2 and γ_3 represent the large-scale spatial trend, while the δ coefficients embody the dependence of log-PM₁₀ concentrations on meteorological variables. Several approaches may be taken for specifying parameters δ , depending on the hypothesised relationship between the variables in question. The most general model is characterised by coefficients that vary in space and time. The specification of time-varying coefficients is particularly useful in embodying nonlinear relationships between pollutant levels and meteorological conditions. Specifying space-varying coefficients implies that the relationship between pollutant level and meteorological conditions changes across space. In order to obtain a model that allows prediction at points where no data are available, some kind of spatial structure has to be imposed on the coefficients if a space-varying relationship is hypothesised. The spatial and temporal dynamics of the regression coefficients can be modelled as mutually dependent or independent. See, for example, Banerjee et al. (2004) for a discussion of space-time varying coefficients models. With regard to the space time modelling of PM₁₀ pollution in the Emilia-Romagna Region, we assume a linear relationship between meteorological variables and PM₁₀, and the same effect is hypothesised at each monitoring site. This is supported by a preliminary analysis indicating that comparable effects at each monitoring site may be postulated (see Cocchi et al., 2006).

The θ_t parameters represent random temporal effects, which in turn account for a residual common dynamic component, once the effect of meteorological conditions has been accounted for. Such parameters are modelled as a random walk process:

$$\theta_t = \theta_{t-1} + \omega_t, \ \omega_t \sim N(0, \sigma_\theta^2) \tag{3}$$

which represents a first-order smoothing non-stationary temporal model. In terms of Dynamic Linear Models, equations (1)-(2) represent the observation equation, while equation (3) is the system equation where θ_t is the state (West and Harrison, 1997). Model (3) is a limiting form of the autoregressive first-order model, and is non-stationary. Within the Bayesian framework, a normal prior distribution is usually assumed for parameter θ_0 , the starting point of the random walk process.

The terms ε_{ts} represent spatially-correlated random effects. They are assumed to follow, at each time t, a multivariate normal distribution with mean vector $\mathbf{0}_{S}$ and $S \times S$ covariance matrix $\sigma_{\varepsilon}^2 \mathbf{\Sigma}$ with ss' entry $\sigma_{\varepsilon}^2 \mathbf{\Sigma}_{ss'} = \sigma_{\varepsilon}^2 exp(-\phi d_{ss'})$. In this way, the logarithm of the correlation is assumed to be a linearly decreasing function of distance $d_{ss'}$ between sites s and s'. The parameter $\phi > 0$ describes the correlation decay rate as a function of distance. This spatial structure is assumed to be constant over time, the underlying assumption being that spatial and temporal processes are separable. A constraint is needed for model identifiability, due to the simultaneous presence of the random temporal effects θ and random spatial effects ε .

4. THE CALIBRATION MODEL

The model described in the previous section is designed for dealing with monitoring networks equipped with homogeneous measuring instruments. In this section we extend the model to cope with data generated by monitoring networks equipped with non co-located heterogeneous measuring instruments. Let S_R and S_E denote the set of monitoring sites equipped with a reference measuring instrument R (an LVG sampler in our application) and with an equivalent non-reference measuring instrument E (a TEOM sampler in our application), respectively. Under the assumption that the reference instrument measures the unknown underlying level with non-systematic errors, equation (1) for each monitoring site $s \in S_R$ at time t can be rewritten as:

$$Y_{t_t} \mid \mu_{t_t}^R, \sigma_s^2 \sim N(\mu_{t_t}^R, \sigma_s^2) \tag{4}$$

where μ_{ts}^{R} represents the "error free" PM_{10} level. The equivalent instrument is supposed to measure the PM_{10} level with some degree of bias. For this reason, we specify equation (1) for each monitoring site $s \in S_{E}$ as:

$$Y_{ts} \mid \mu_{ts}^{E}, \sigma_{s}^{2} \sim N(\mu_{ts}^{E}, \sigma_{s}^{2}) \tag{5}$$

where μ_{ts}^{E} represents the PM₁₀ level as measured by the equivalent measuring instrument, free from random error σ_{s}^{2} . The crucial feature of the model is that the mean produced by the equivalent measuring instrument can be expressed as a function (indexed by a set of parameters Ψ^{*}) of the true underlying pollution level, that is, the level that would have been measured by the reference sampler:

$$\mu_{ts}^{E} = f(\mu_{ts}^{R}; \Psi^{*}). \tag{6}$$

The problem of calibration thus comes down to the specification of the form of the functional relationship (6) and to the consequent parameter estimation.

However, the relevance of a calibration study does not lie in the estimation of parameters allowing the transformation of reference values into equivalent values. The focus is rather on the estimation of parameters which transform non-reference measures into reference measures according to the inverse function:

$$\mu_{tt}^{R} = f^{-1}(\mu_{tt}^{E}; \Psi). \tag{7}$$

When the calibration function $f(\cdot)$ is linear, equation (6) becomes $\mu_{ls}^E = \alpha^* + \beta^* \mu_{ls}^R$, where $\Psi^* = (\alpha^*, \beta^*)$. Parameters μ_{ls}^R are modelled as parameters μ_{ls} in (2) $\forall s \in (S_R \cup S_E)$. Parameters α^* and β^* represent the additive and multiplicative bias characterising the equivalent measurement instrument. In this parameterisation, the E instrument's expected measurements are expressed as a linear function of the expected reference measurements at the same site. Equation (7) becomes: $\mu_{ls}^R = \alpha + \beta \mu_{ls}^E$, where $\Psi = (\alpha, \beta)$ and parameters μ_{ls}^E are modelled as in (2) $\forall s \in (S_R \cup S_E)$. If the model is parameterized as in (6), equation (2) expresses the mean value at point s on day t on the reference measuring instrument scale. If the model is re-parameterised using α and β as in (7), then (2) expresses the mean value on the equivalent measuring instrument scale. The measuring instruments are treated symmetrically and the parameterisation can be designed in order to estimate the calibration parameters in question.

The model hierarchy is completed by the prior specification for model parameters. According to an approximately non-informative criterion, proper, albeit flat, prior distributions have to be specified. Normal independent priors N(0,1000) are assumed for each component of the coefficient vectors $\boldsymbol{\delta}$ and $\boldsymbol{\gamma}$. Independent small parameters' inverse Gamma distributions IG(0.01,0.01) are specified for the variance parameters σ_s^2 , s=1,...,S, while uniform prior distributions U(0,10) are specified for the second-level standard deviations σ_{θ} and σ_{ε} . A uniform distribution U(0,2) is assumed for $\boldsymbol{\phi}$: this results in a prior belief for the spatial correlation ranging from 0.13 to 1 at a distance of 1 km, and from 0 to 1 at the maximum distance of 250 km. Our choice of a prior for parameter θ_0 is a normal distribution whose mean is equal to the observed mean at 31st December 1999, and whose variance is equal to 10, in order to specify a fairly vague prior.

If the analyses are performed on the log-scale, the "1.3 rule" for the calibration of the TEOM measurements is a special case of the proposed model, with $\alpha = log(1.3)$ and $\beta = 1$. On the original scale, this implies a linear relationship, with the intercept term equal to 0. The proposed model with a linear calibration function implies, on the original scale, the non-linear relationship $PM_{10,LVG} = exp^{\alpha}PM_{10,TEOM}^{\beta}$.

Since different relationships between reference and equivalent measures may be hypothesised, certain adaptation criteria under the different specification of (6) have to be used in order to choose the most suitable functional relationship. A widely used adaptation criterion within the Bayesian framework is the Deviance Information Criteria (DIC; Spiegelhalter *et al.*, 2002), a Bayesian generalisation of the Akaike Information Criterion that takes account both of model fitting and of model complexity. When comparing model performances using DIC, the chosen model is the one with the lowest DIC value.

5. DISCUSSION AND RESULTS

We estimated the previously-suggested model for the Emilia-Romagna Region's monitoring network, on a set (S_R) of 11 monitoring sites equipped with LVG samplers and one monitoring site equipped with a TEOM sampler. In order to check the appropriateness of the linear calibration function, we specified two different calibration functions, the first linear and the other quadratic. In terms of DIC, the linear function is preferable; furthermore, the MCMC sampling algorithm displays a degree of inefficiency when estimating the quadratic function's parameters.

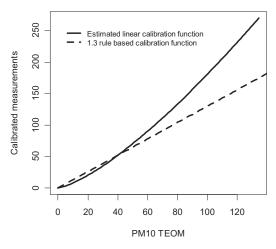
Samples from the parameters' posterior distributions were obtained using an MCMC algorithm as implemented in the WinBUGS software following a multichain approach. Convergence has been checked by graphical examination of the trace plots of the chain's sample values versus iterations, and of the autocorrelation plot for each chain. Moreover, we computed the Gelman-Rubin convergence statistic as modified by Brooks and Gelman (1998). Posterior summaries are obtained by sampling 20,000 post-convergence samples after a burn-in of 30,000. Table 1 summarises the calibration parameters' posterior distributions.

TABLE 1

Posterior summaries for the calibration parameters

Parameter	Mean	2.5th perc.	97.5th perc.
α	-1.076	-1.177	-0.991
β	1.362	1.336	1.392
$\exp(\alpha)$	0.342	0.308	0.371

According to the parameters' posterior mean, the estimated relationship is non-linear ($\beta \neq 1$) and convex. This suggests that TEOM samplers tend to overestimate low levels, and to heavily underestimate high levels, of PM₁₀. According to this result, the 1.3 factor mainly fails to correctly calibrate TEOM measurements in the case of high PM₁₀ levels. As shown in Figure 2, underestimation of the true level can be serious in the case of acute pollution events, and in general during the winter, when pollution levels measured by LVG samplers are very often higher than 80 μ g/m³. The inadequacy of the "1.3 rule" has been checked by estimating the proposed model under the hypotheses that the calibration parameters are set at $\alpha = \log(1.3)$ and $\beta = 1$: the DIC for this model is 2140, compared with a value of 1980 obtained using the linear calibration function.



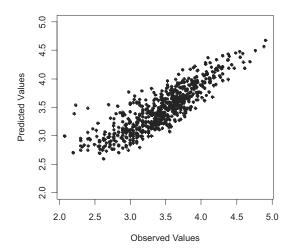


Figure 2 – The estimated calibration function and the "1.3 rule" calibration function.

Figure 3 – Predicted vs observed TEOM measurements in the out of sample site 13.

Model effectiveness in spatial prediction has been checked using an off-sample monitoring site (site 13 in Figure 1) equipped with a TEOM sampler: both TEOM and calibrated measurements have been predicted for this site. Figure 3 shows the scatter plot of observed versus predicted values. The correlation between predicted and observed TEOM measurements is r=0.87, showing the satisfactory performance of the proposed model. It is worth noting that the prediction is satisfactory despite the fact that the out of sample monitoring site (site 13) is at a considerable distance from the monitoring sites utilised for model estimation. This is favoured by the homogeneity of the pollutant-generating process in the Emilia Romagna Region, which ensures the strong spatial representativeness of each monitoring site. An efficient calibration rule is important when evaluating particulate matter pollution levels for two main reasons: first of all, it produces reliable population exposure levels in ecological regression studies designed to evaluate the effect of air pollution on the public's health; secondly, it enables air quality to be evaluated in order to assess if environmental standards are being met. Since environmental standards for PM₁₀ are set according to LVG sampler, using noncalibrated TEOM measurements may underestimate the number of exceedances, with the consequence that a site could be deemed to comply with requirements when in fact it does not, simply because its measuring instrument is biased. Table 2 below shows the environmental standards in force during the study period in question.

TABLE 2

Environmental standards for PM_{10} levels ($\mu g/m^3$)

Year	2000	2001	2002
Threshold	75	70	65
Annual mean	48	46.4	44.8

A site is considered to comply with such standards if the mean daily concentrations does not exceed the threshold reported in the first row of Table 2 on more than 35 days per year, and if the annual mean does not exceed the values reported in the second row of table 2. Since air quality assessment is based on the number of exceedances, a model that can produce reliable predictions in space and time is useful since it enables scientists to obtain complete time series (that is, free from missing observations).

When evaluating the annual mean and the number of exceedances, missing observations have to be imputed since the period during which there are no recorded observations heavily influences results: for example, if missing observations are concentrated during winter, the annual mean and the number of exceedances may be strongly underestimated if only observed data are considered. For this reason, we are now going to focus on the evaluation of compliance with environmental standards once missing observations have been imputed by the estimated model.

Calibration is important when evaluating air quality at sites equipped with a TEOM sampler, as this tends to underestimate the number of exceedances per year. In order to see this, we focus on the number of exceedances at site 12, and compare:

- the number of exceedances and the annual mean predicted on the TEOM scale after imputation of missing values using the mean of their posterior distributions;
- the number of exceedances and the annual mean predicted on the LVG scale once missing values have been imputed using the mean of their posterior distributions after calibration using the "1.3 rule" and our model;
- the number of exceedances and the annual mean predicted at site 4 once missing values have been imputed using the mean of their posterior distributions.

Comparison with site 4 is of interest because of its proximity to site 12. Since the distance between the two sites is only 3 km, it is reasonable to expect a comparable number of exceedances and similar annual means at the two sites. However, site 12 is located in a background urban area whereas site 4 is located in a densely-populated area. They may be compared once the difference between the site types has been taken into account: using model (2): this was done simply by predicting data at site 4 as if it was a background site, that is by setting Z_4 =1.

TABLE 3

Number of exceedances and predicted annual mean

Year		2000	2001	2002	2000-2002
Site 12					
TEOM	Exceedances	14	8	15	37
	Mean	40.7	32.8	32.8	35.46
1.3 rule	Exceedances	49	28	41	118
	Mean	52.9	42.6	42.7	46.1
Calibrated Values	Exceedances	68	37	43	148
	Mean	53.3	39.8	40.2	44.5
Site 4					
LVG	Exceedances	68	36	42	146
	Mean	53.2	39.2	39.3	43.92

As shown in Table 3, non-calibrated TEOM measurements tend to heavily underestimate both the annual mean and the number of exceedances. The "1.3 rule" predicts a substantially lower number of exceedances than were predicted by the displaced calibration rule in the year 2000 when PM₁₀ levels were higher than in subsequent years. As the mean pollution level drops, the difference between predicted exceedances is reduced. As regards mean levels, the "1.3 rule" tends to produce a higher annual mean for years 2001 and 2002 than the displaced calibration rule does. This is because the "1.3 rule" overestimates the low pollution levels that were often observed in 2001 and 2002. Proof of the proposed model's effectiveness in ascertaining whether environmental standards are being met is provided by the comparison between the number of exceedances and the forecast annual means at site 4 (equipped with an LVG sampler), and at site 12 after displaced calibration of the TEOM measurements. In fact, since the sites are very close, we expect the air quality at the two sites to be comparable. Table 3 shows that, after the displaced calibration, the air quality at the sites in question is very similar, as we would have expected.

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RIASSUNTO

Calibrazione di misure eterogenee di PM₁₀ mediante l'uso di modelli spazio-temporali

Le reti di monitoraggio del PM₁₀ sono spesso caratterizzate da eterogeneità degli strumenti di misurazione. E' noto che alcuni di questi strumenti sottostimano il valore vero dell'inquinante. Nel presente articolo si propone un modello gerarchico spazio-temporale bayesiano per la calibrazione di misure effettuate da strumenti diversi da quello di riferimento, sfruttando l'informazione derivante da misure rilevate da strumenti di riferimento non co-locati nello spazio.

SUMMARY

Displaced calibration of PM_{10} measurements using spatio-temporal models

PM₁₀ monitoring networks are equipped with heterogeneous samplers. Some of these samplers are known to underestimate true levels of concentrations (non-reference samplers). In this paper we propose a hierarchical spatio-temporal Bayesian model for the calibration of measurements recorded using non-reference samplers, by borrowing strength from non co-located reference sampler measurements.