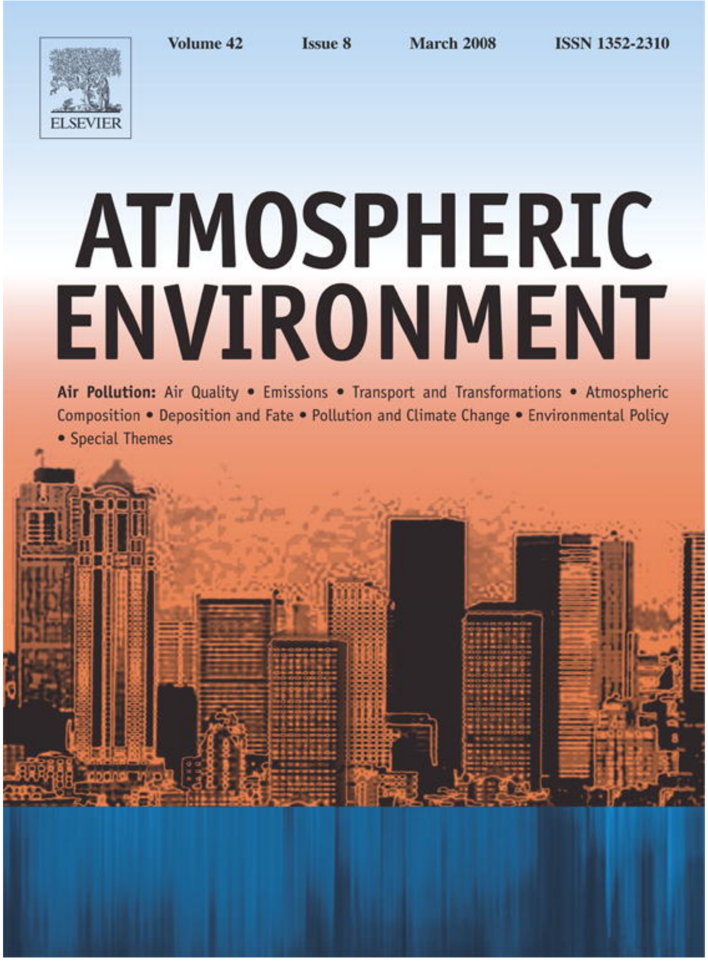
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The effect of salting with magnesium chloride on the concentration of particular matter in a road tunnel

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Abstract

This work presents an analysis of the effect of dust treatment with a magnesium chloride solution on the concentration of airborne particulate matter inside a road tunnel. During the winter 2004/2005, the road inside the tunnel of Strømsås was salted 43 times with magnesium chloride solution, of which 27 times with the amount 20 $\mathrm{g\,m^{-2}}$ and 16 with 40 $\mathrm{g\,m^{-2}}$. In addition, the road was swept 11 times and washed twice. Simultaneously, one recorded the concentration of PM_{10} and $PM_{2.5}$ hourly. The model used in the analysis is a generalised additive model (GAM) on log-scale, having traffic counts, diverse meteorological conditions and the pollution reducing actions as predictor variables.

The analysis revealed no clear effect from sweeping and washing. The impact of the dust binding medium on the concentrations of particulate matter is, however, clearly propitious. As one would expect, it is largest immediately after applying the magnesium chloride, and diminishes steadily afterwards. The duration of the effect is estimated to 10 days, but with a rather large uncertainty (95% confidence intervals between 3 and 16 days). We estimated an effect (reduced pollution level) of 70% on the concentration of the coarse particles PM_{10}–$PM_{2.5}$ and 56% on the concentration of PM_{10}. The estimated effect on the fine particles $PM_{2.5}$ is a modest 17%, and barely significant.

Keywords: Air quality modelling; Urban air quality; Dust treatment; Road dust; Generalised additive models

1. Introduction

Traffic-related air pollution has caused severe health problems and bothers over the last decades (Brunekreef and Forsberg, 2005; Næss et al., 2006). As a result, the focus on reducing and preventing air pollution has been massive, and several measures have been initiated. One category of actions seeks to cut the emission of pollutants. For instance, the use of studded tyres, abrading asphalt-paved roads, is restricted by a charge in Norway. Another category of measures aims to prevent particles on the road surface from whirling up into the air (suspension). Such actions include reducing speed limits, sweeping and washing the roads and using dust binding materials on the road surface.

Magnesium chloride solution (20% $MgCl_2 \cdot 6H_2O$ and 80% water) has been used against dust suspension on gravel roads for a long time. Since the end of the 1990s it has also been applied on some asphalt-paved roads in Norway, both in road tunnels and on open-air roads. The aim of this paper is to document the dust binding effect of magnesium chloride on the

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concentration of airborne particulate matter inside a road tunnel. During the winter 2004/2005 for a period of 26 weeks the nearly 4 km long bi-directional "Strømsås tunnel" in the city of Drammen, Norway, was salted 43 times with magnesium chloride, of which 27 times with the amount 20 g m^{-2} and 16 with 40 g m^{-2}. The road surface was swept 11 times and washed twice during the same period. Simultaneously, the concentrations of PM_{10} and $PM_{2.5}$, as well as traffic volume and meteorological conditions inside the tunnel, were monitored. We have analysed these data using a generalised additive model (GAM) (Hastie and Tibshirani, 1990) on log-scale. The concentration of the three pollution components PM_{10}, $PM_{2.5}$ and PM_{10}–$PM_{2.5}$ was response variables and traffic volume, meteorological variables and pollution reducing measures predictor variables. We estimated separate models for each of the pollution components. Previous work (Aldrin and Hobæk Haff, 2005) has proved this to be a suitable model for such an application. We found that applying magnesium chloride had a rather large positive effect on the coarse particles and a much smaller, barely significant, positive effect on fine particles.

Similar, but less extensive, experiments using magnesium chloride have been conducted in other locations in Norway the last few years. Two of these, conducted in tunnels, concluded with a positive effect (approximately 50% reduction in the PM_{10} level), one in Trondheim (Værnes, 2003) and one in Oslo (Tønnesen, 2006). Beneficial results (roughly 25% reduction) were also reported from one experiment on open-air streets in Trondheim (Berthelsen, 2003), whereas Aldrin and Steinbakk (2003) did not find any significant effect in another open-air experiment on motorways in Oslo with speed limit 80 km h^{-1}. Another successful open air experiment, using a calcium magnesium acetate (CMA) solution, documents a 29–43% reduction of the concentration of PM_{10} in Klagenfurt, Austria (Hafner, 2007). However, none of these studies have been published in scientific journals. In fact, we have found only one article in peer reviewed journals which deals with dust binding materials on paved roads. Norman and Johansson (2006) reported that application of CMA on (open-air) roads in Stockholm, Sweden, reduced the level of PM_{10} with 35%.

The outline of the paper is as follows. Section 2 exhibits the data and experimental design. The methods used in the analysis are presented in Section 3. The corresponding results are summarised in Section 4 followed by a discussion and conclusion in Section 5.

2. Data and experimental design

The data set consists of concentrations of particulate matter (hourly average), diverse meteorological measurements (hourly average) and traffic counts (hourly aggregated data) from the Strømsås tunnel, just outside Drammen. This is a 4 km long bi-directional tunnel through which 6000 vehicles pass daily. Concentrations of PM_{10} and $PM_{2.5}$ were recorded with the R&P 1400 TEOM monitor during the period from 18 October 2004 to 17 April 2005, which corresponds to 26 weeks. In addition, we have computed the concentration of coarse particles as the difference PM_{10}–$PM_{2.5}$. Traffic counts are divided into light (length 5.5 m or shorter) and heavy (longer than 5.5 m) vehicles. These hourly counts are from the same time period as the concentrations of particulate matter. Corresponding meteorological data were also measured inside the tunnel with a Vantage PRO. The recorded variables are the temperature, the relative humidity, the wind direction, the wind velocity, the dew point and the atmospheric pressure. Their units are given in Table 1.

The tunnel is equipped with ventilation fans. These are turned on when the exhaust level is high, blowing from east to west. This obviously affects the concentration of particulate matter inside the tunnel, especially on dry and dusty days. Unfortunately, the use of the fans has not been logged. Thus, the effect of ventilation cannot be included in the model. Furthermore, about 30% of drivers changed from summer tyres to studded tyres in the beginning of the study period and changed back to summer tyres again at the end of the period. The use of studded tyres is known to increase the concentration of particulate matter (Bartonova et al., 2002;

Table 1
Meteorological variables recorded in the tunnel

Variable	Unit
Temperature	Degrees Celsius
Relative humidity	%
Wind direction	Degrees, 0 = North
Wind velocity	m s^{-1}
Dew point	Degrees Celsius
Atmospheric pressure	mmHg

Gustafsson et al., 2005). However, as the data series are shorter than one year, and there is no reference period without use of studded tyres, we will not be able to discern this effect from other time-varying effects.

The aim of this paper is to investigate the effect of applying magnesium chloride to the road surface in the tunnel. Two different amounts of a 20% magnesium chloride water solution $MgCl_2 \cdot 6H_2O$, namely 20 and $40\,g\,m^{-2}$, have been used in the study, the former 27 times and the latter 16. In 26 of the cases, the time interval between salting events was shorter than three days, in 13 cases it was three to four days, whereas it was five days or more in only four cases. The salt was dispersed by a salt spreader (Nido Stratos B70-30), covering the whole surface, including the carriage way, the verge and the lower 10–20 cm of the tunnel walls, with salt. To avoid confounding rush time cycles and salting, the salt treatment was performed randomly in one of the four time intervals: 0–4 a.m., 8–10 a.m., 1–2 p.m. and 6–7 p.m. In addition, the carriage way and the verge have been swept 11 times and washed twice in the same winter period. Sweeping (with a Beam S7000) was always carried out between 12 p.m. and 4 a.m. and washing with a high pressure water cleaning system between 9 p.m. and 4 a.m. Salting is the cheapest of these treatments, with a cost of about 130 Euro per kilometre. The cost of sweeping is about 210 Euro per kilometre, and washing is even more expensive.

The concentration of coarse particles (PM_{10}–$PM_{2.5}$) from week number 10 to week number 13, 2005 is plotted on log-scale in the two upper panels of Fig. 1. (Corresponding values of PM_{10} and $PM_{2.5}$ are found in Fig. 5.) Gaps in the line correspond to periods of missing data. The dotted vertical lines indicate the last hour of each day. Pollution reducing actions are marked with vertical lines of different colours. Sweeping is indicated by yellow lines, and salting with 20 and $40\,g\,m^{-2}$ by purple and blue, respectively. During the selected four weeks, the road was never washed. Sometimes, the application of magnesium chloride is followed by an immediate visible pollution reduction, such as on 21 March 2005, but not always. The particulate matter level is obviously influenced by other conditions as well. Thus, it is necessary to correct for variables, like traffic, weather, washing and sweeping, to be able to detect a potential dust binding effect of magnesium chloride.

The two medium panels of Fig. 1 show the number of light vehicles passing through the tunnel each hour during the same four weeks as in the two upper panels. As expected, the traffic counts exhibit distinct diurnal and hebdomadal patterns, except for the period between Thursday 17 March and Tuesday 29 March, when they are affected by the easter holidays. Corresponding measurements of relative humidity are displayed in the two lower panels of Fig. 1. The humidity is mostly rather stable, but has a few peaks.

3. Methods

3.1. Basic model

We have modelled each pollution component (PM_{10}, $PM_{2.5}$ and PM_{10}–$PM_{2.5}$) with a GAM (Hastie and Tibshirani, 1990) on log-scale, having traffic counts, meteorology, time of day, a long-term time trend and the pollution reducing measures as predictor variables, similar to the models presented in Aldrin and Hobæk Haff (2005). The model is

$$\log(y_t) = s_1(x_{1t}) + \cdots + s_p(x_{pt}) + \varepsilon_t, \tag{1}$$

where y_t is the concentration of PM_{10}, $PM_{2.5}$ or PM_{10}–$PM_{2.5}$ at time (h) t, the x_{it}'s are the predictor variables and ε_t is the residual, i.e. the part of $\log(y_t)$ that the model does not explain. Furthermore, the $s_i(\cdot)$'s are unknown, smooth functions that must be estimated from the data. Aldrin and Hobæk Haff (2005) showed that such a model is well suited for this application. On original scale, the effects are multiplicative, and the model can be written as

$$y_t = S_1(x_{1t}) \cdots S_p(x_{pt}) \cdot E_t, \tag{2}$$

where $S_i(\cdot) = \exp(s_i(\cdot))$ and $E_t = \exp(\varepsilon_t)$.

As mentioned earlier, traffic volumes are divided into light and heavy vehicles. The meteorological variables are the ones presented in Table 1. Furthermore, the hour of day is included as a predictor variable to take into account the diurnal variation that is not explained by variables such as traffic counts and temperature. We have also included the time since the start of the data period as a predictor variable. It is supposed to handle long-term variation and potential seasonality.

The pollution preventing measures are modelled as four separate effects. In an extended analysis (not shown here) we investigated potential interactions between them, but found none to be significant. Therefore, interactions are not considered further in this paper. The corresponding variables are specified as follows. Sweeping is represented by the time

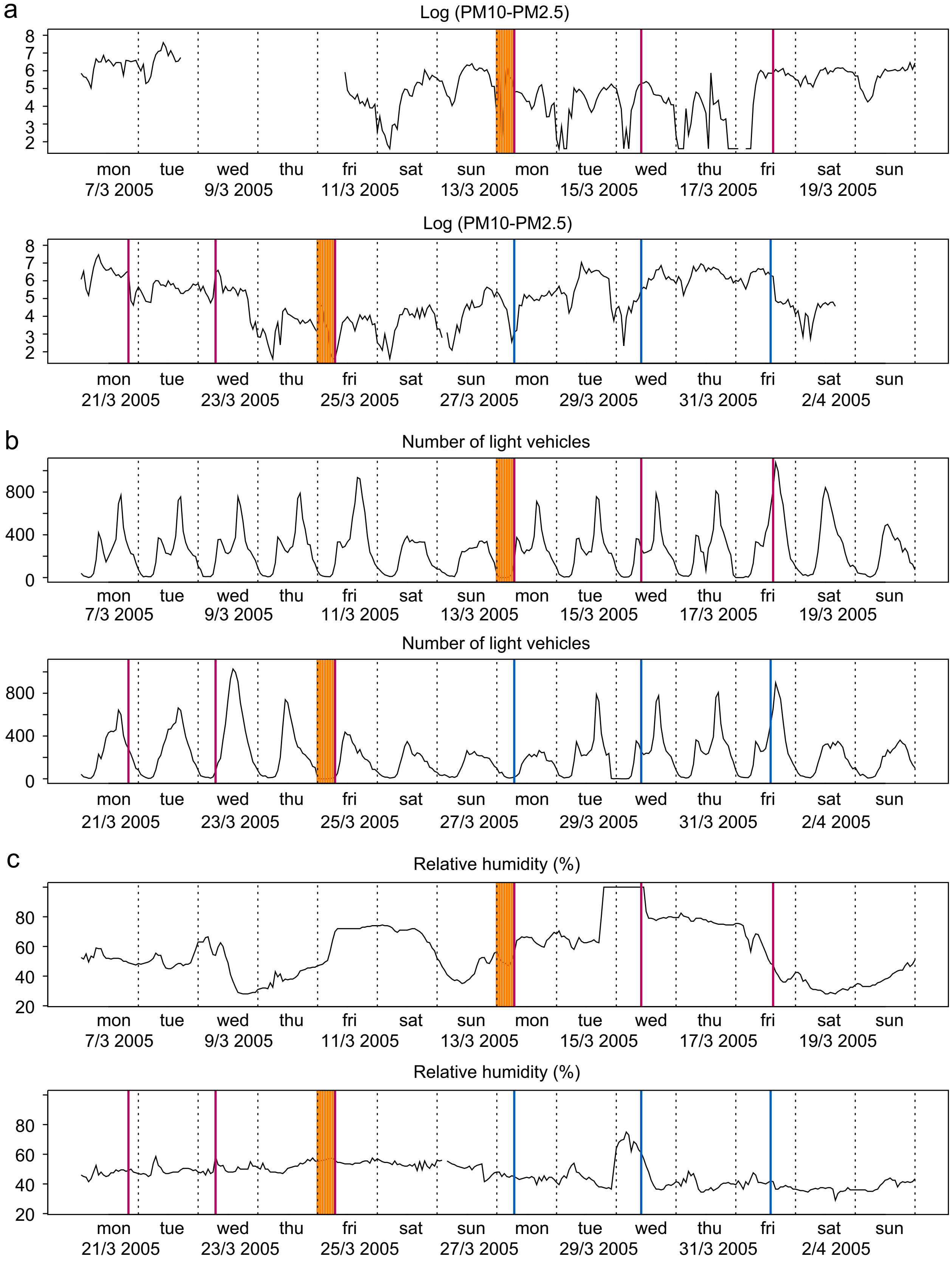

Fig. 1. Hourly values of (a) the natural logarithm of PM_{10}–$PM_{2.5}$, (b) the number of light vehicles (equal to or shorter than 5.5 m), and (c) the relative humidity during the weeks 10–13 2005. Vertical lines mean: yellow, sweeping; purple, salting with 20 mg^{-2}; blue, salting with 40 mg^{-2}; dotted black, last hour of each day.

since the road was swept the last time, up to a maximum θ^{sweeping}. After that, it is assumed to have no more effect. Note that θ^{sweeping} is the *maximum possible* duration of the sweeping effect, but since the *s*-functions in model (1) are non-linear and flexible, the *actual duration* may be shorter. Moreover, the impact of the previous sweeping is presumed to be negligible when the road is swept once more. This may be expressed as

$$\text{Sweeping} = \min(\text{time since the last sweeping}, \theta^{\text{sweeping}}).$$

The washing variable is defined similarly, as

$$\text{Washing} = \min(\text{time since the last washing}, \theta^{\text{washing}}),$$

where θ^{washing} is the assumed maximum possible duration of the washing effect.

One has applied two different amounts of magnesium chloride, namely 20 and 40 g m^{-2}. Hence, we include two salting variables in the model, representing each of the amounts. They resemble the sweeping and washing variables. However, they are strongly related and must be handled as such. More specifically, we assume that the effect of a previous salting ceases as a new one, with either amount, is performed. This may be written as

$$\text{Salting20} = \begin{cases} \min(\text{time since the last salting}, \theta^{\text{salting}}) & \text{if salting with } 20\,\text{g m}^{-2}, \\ \theta^{\text{salting}} & \text{if salting with } 40\,\text{g m}^{-2}, \end{cases}$$

$$\text{Salting40} = \begin{cases} \min(\text{time since the last salting}, \theta^{\text{salting}}) & \text{if salting with } 40\,\text{g m}^{-2}, \\ \theta^{\text{salting}} & \text{if salting with } 20\,\text{g m}^{-2}, \end{cases}$$

where θ^{salting} is the maximum possible duration of the salting effect, after which the impact of salting is assumed to be negligible.

The models were estimated by maximum likelihood, using the software package S-Plus (version 7.0.6, Insightful Corporation, Seattle, WA). The flexibility of each *s*-function is controlled by specifying its number of degrees of freedom. We have fixed this number to four (the default in S-Plus) for all *s*-functions but two. For the hour of day, it was set to 10, and for the time since the start of the data period, it was fixed at 20, allowing for more flexibility in the functions. These choices of degrees of freedom are motivated by a cross-validation experiment with similar data in Aldrin and Hobæk Haff (2005).

3.2. *Simplified models*

In order to facilitate interpretation and impartment to potential users, we will simplify the model by linearising the effect of salting (on log-scale). This is done by assuming that salting (with both amounts) has an immediate effect that decreases linearly throughout the duration θ^{salting}. The parameter θ^{salting} is now interpreted as the *actual* duration of the effect, as opposed to model (1) where it was interpreted as the *maximum possible* duration. The new model is

$$\log(y_t) = s_1(x_{1t}) + \cdots + s_{p-2}(x_{p-2,t}) + \beta_{20} \cdot \text{Salting20}_t + \beta_{40} \cdot \text{Salting40}_t + \varepsilon_t, \tag{3}$$

where $x_{1t}, \ldots, x_{p-2,t}$ are the remaining predictor variables, including sweeping and washing. Note that the salting variables Salting20 and Salting40, defined in Section 3.1, depend on the duration θ^{salting}, which is treated as an unknown parameter. The coefficients β_{20} and β_{40}, the *s*-functions and the duration θ^{salting} are estimated by maximum likelihood. This is done simultaneously for the three pollution components, assuming the same duration θ^{salting} for all three components and for both amounts of magnesium chloride. Technically this is done by estimating the model (3) for each of three pollution components for a fixed value of θ^{salting}. Then, θ^{salting} is varied over a grid of reasonable values, to find the one that maximises the total log-likelihood

$$l_{\text{tot}} = l_{\text{PM}_{10}-\text{PM}_{2.5}} + l_{\text{PM}_{10}} + l_{\text{PM}_{2.5}}, \tag{4}$$

where the *l*'s on the right-hand side are the log-likelihoods for each pollution component.

We simplify the model (3) further, replacing the two salting effects (corresponding to the two amounts) by a common effect. This model is

$$\log(y_t) = s_1(x_{1t}) + \cdots + s_{p-2}(x_{p-2,t}) + \beta \cdot \text{Salting}_t + \varepsilon_t, \tag{5}$$

where

$$\text{Salting} = \min(\text{time since the last salting}, \theta^{\text{salting}})$$

and β is the common effect on log-scale. The duration of the salting effect, θ^{salting}, is estimated in the same way as for model (3).

The immediate common effect of salting (1 h after it was performed), relative to the level after it has dissipated, is given by $\beta \cdot (1 - \theta^{\text{salting}})$ on log-scale. However, we are more interested in its effect on the original scale. We define this as the relative decrease in pollution level immediately after salting, compared to the corresponding pollution level without salting. In percent this is given by

$$\text{Immediate common effect of salting} = 100(1 - \mathrm{e}^{\beta \cdot (1-\theta^{\text{salting}})}). \tag{6}$$

The two separate effects of salting with different amounts are computed in the same way.

3.3. Assessment of parameter uncertainty

So far we have implicitly assumed that the residuals ε_t in (1), (3) and (5) are uncorrelated in time. However, this is an unrealistic assumption for our time series data. The estimates of the β-coefficients are unbiased, even though the autocorrelations are ignored (see for instance Mardia et al., 1979, Chapter 6.6), but their uncertainty may be severely underestimated. Furthermore, the uncertainty estimates provided by standard software such as S-Plus do not take into account that the duration of the effect, θ^{salting}, is unknown.

We calculate the uncertainty of the parameters of interest by a semi-parametric variant of the bootstrap (Efron and Tibshirani, 1993), taking into account autocorrelation as well as the uncertainty in θ^{salting}. First the models are estimated from the data, and the empirical residuals are calculated. Then new bootstrap data sets are generated by resampling the residuals using a stationary block bootstrap (Politis and Romano, 1994; Lahiri, 2003), approximately preserving the autocorrelation. The model is re-estimated for each bootstrap data set (including re-estimation of the duration), yielding bootstrap replicates of the parameters of interest. Finally, confidence intervals are estimated by the percentile interval method (Efron and Tibshirani, 1993, Chapter 13.3). Details on the bootstrap procedure are found in Appendix A.

4. Results

4.1. Results from the basic model

Based on the data from Section 2, we have estimated model (1) for each of the three particulate matter variables PM_{10}, $PM_{2.5}$ and PM_{10}–$PM_{2.5}$. The maximum possible durations of the sweeping and washing effects (θ^{washing} and θ^{sweeping}) are set to 40 days, whereas the maximum possible duration of the salting effect (θ^{salting}) is set to 20 days. We chose to set the maximum possible durations of sweeping and washing effects longer than for salting, since sweeping and washing potentially remove the dust completely.

Fig. 2 shows the estimated s-functions on log-scale for the coarse particles PM_{10}–$PM_{2.5}$. The 95% confidence intervals shown here are the standard intervals supplied by S-plus, ignoring the autocorrelation in the residuals. Hence, the true uncertainties are larger. The time since the start of the data period and the time since washing, sweeping and salting have unit days in this figure, but with an hourly resolution (1/24 day).

The effect of salting with $20\,\mathrm{g\,m^{-2}}$ is displayed in the lower left corner. The leftmost point of the curve, showing the effect immediately after salting, should be compared to the rightmost point, which reflects the reference level without salting or after the effect has diminished. The curve increases from the left to the right, indicating that salting with $20\,\mathrm{g\,m^{-2}}$ has an immediate effect that diminishes with the time since it was performed. The difference between the extremities is about one (on the log-scale), which means that salting can reduce the concentration of PM_{10}–$PM_{2.5}$ by about $\frac{1}{3}$ (since $\exp(-1) \approx \frac{1}{3}$). The effect appears to level off after approximately 10 days or more. The corresponding impact from salting with $40\,\mathrm{g\,m^{-2}}$ (shown in row four, column two of Fig. 2) is less clear, but seams to be similar to the effect of half the amount. The odd shape of the curve is due to the lack of data for durations between eight and 20 days after salting (indicated by the corresponding lack of black tick marks at the bottom of the panel).

The effect of sweeping, displayed in row three column three of Fig. 2, is difficult to interpret. The estimated curve suggests that the maximum effect occurs after 20–30 days, which is highly unlikely. Washing (shown in row three column four), on the other hand, appears to have no impact. However, we cannot exclude that sweeping and washing have

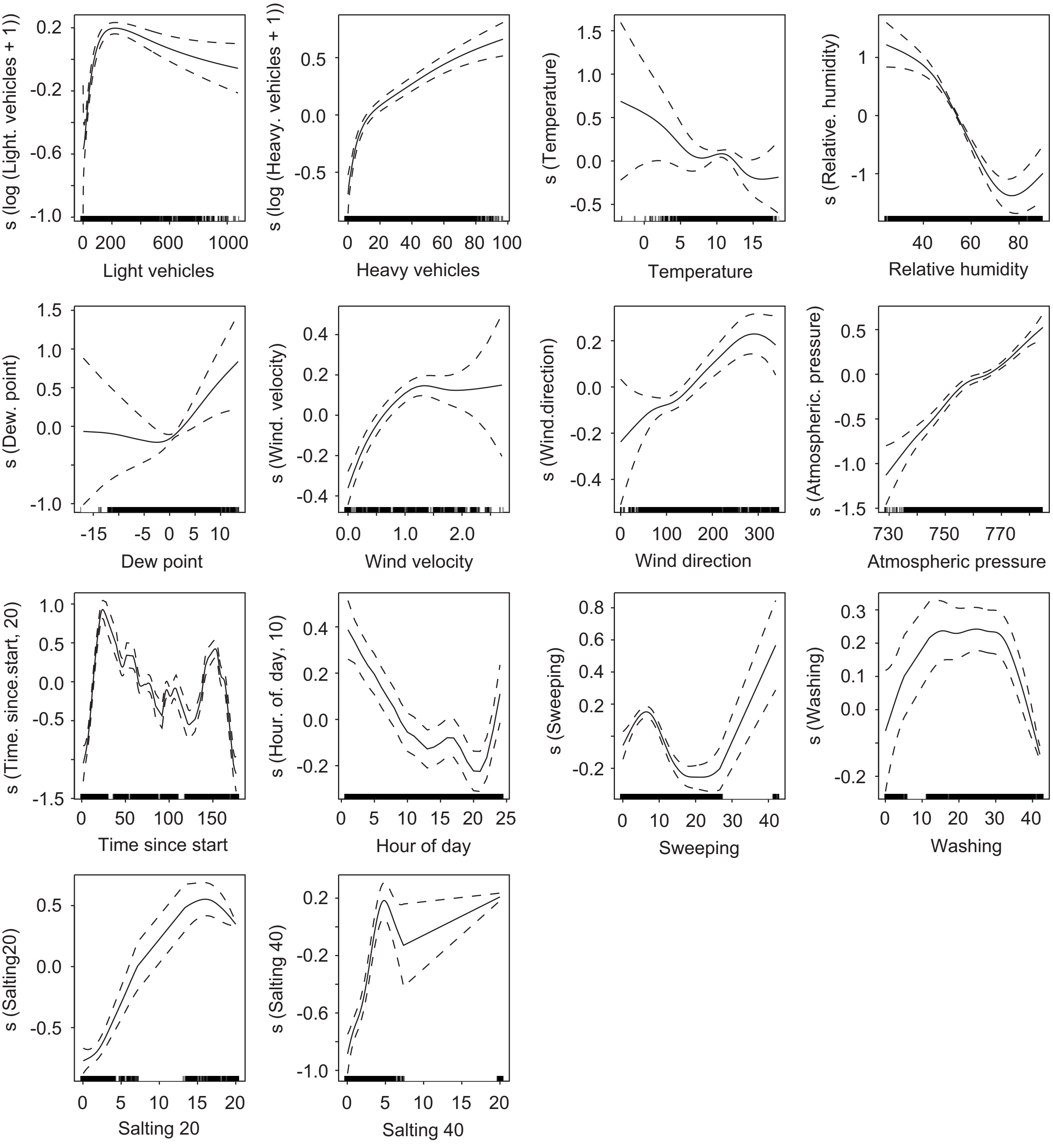

Fig. 2. Estimated *s*-functions from model (5) fitted to the natural logarithm of PM_{10}–$PM_{2.5}$. The 95% confidence intervals have been constructed without considering the autocorrelation in the residuals. The true confidence intervals are therefore larger. The black tick marks at the bottom inside of each panel indicate the value of the explanatory variable at each data point.

an effect on particulate matter concentrations. Hence, we will keep them in the models to adjust for possible confounding with salting, but will not interpret their effects any further.

The remaining predictor variables are clearly important, especially traffic, relative humidity, temperature and dew point. Concentrations of particulate matter obviously rise with increasing traffic counts, in particular up to 200 light vehicles and 10 heavy ones. Increasing temperature and relative humidity induce decreasing PM-levels, while the dew point has the opposite effect. The remaining meteorological variables, wind velocity, wind direction and atmospheric pressure have some impact on the concentrations of particulate matter, but less so than the others. The curves for the time since the start and hour of day reveal both a long-term and diurnal variation, respectively, that is not explained by the rest of the model, among others a reduced pollution level around 8 p.m. The large increase and drop at the start and end, respectively, of the curve for the time since the start may be explained by the

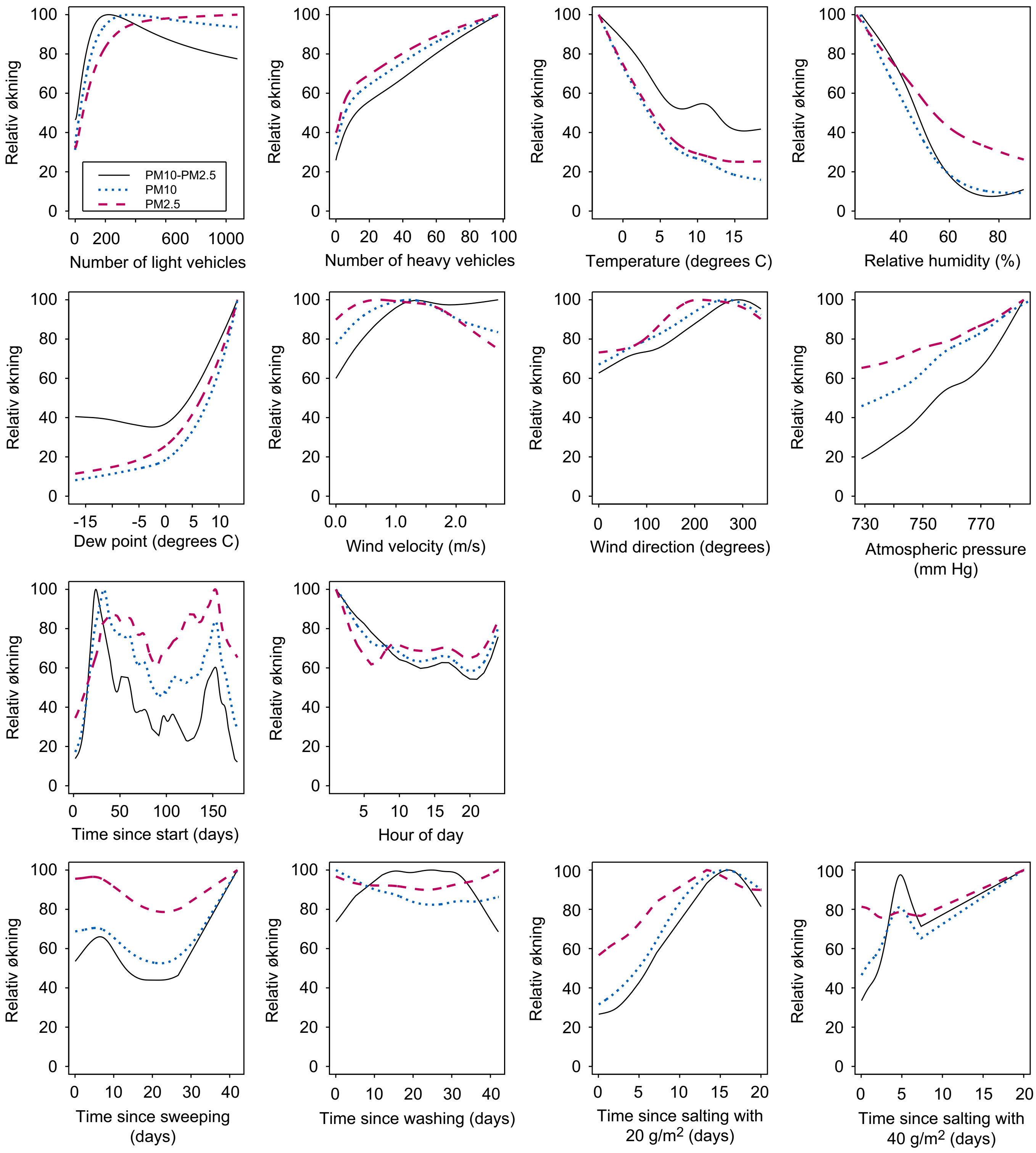

Fig. 3. Estimated *S*-functions (i.e. on original scale) from model (5) for PM_{10}–$PM_{2.5}$, PM_{10} and $PM_{2.5}$. The maximum value of each curve is set to 100.

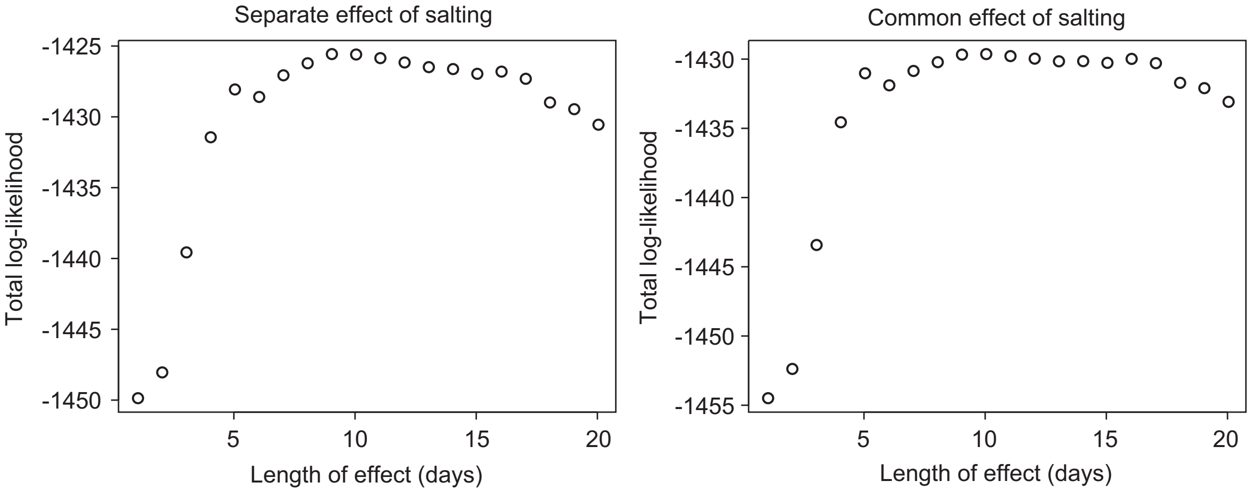

Fig. 4. Total likelihood as a function of the duration θ^{salting} of the salting effect. Left panel: separate effects for each amount of magnesium chloride based on model (3). Right panel: common effect for both amounts of magnesium chloride based on model (5).

Table 2
Estimates and confidence intervals of duration (in days) and magnitudes (in %) of immediate separate effects of salting with the two amounts, based on model (3)

Pollution component	Duration of the effect		Effect 20 g m^{-2}		Effect 40 g m^{-2}		Difference between 20/40 effects	
	Est.	95% conf. int.	Est.	95% conf. int.	Est.	95% conf. int.	Est.	95% conf. int.
PM_{10}–$PM_{2.5}$	9	(3, 17)	70	(47, 90)	67	(39, 88)	3	(−12, 20)
PM_{10}	9	(3, 17)	63	(37, 77)	48	(17, 68)	15	(4, 28)
$PM_{2.5}$	9	(3, 17)	29	(10, 40)	8	(−13, 25)	21	(8, 35)

change from summer tyres to studded tyres and back again.

We have transformed the estimated *s*-functions on logarithmic scale back to the corresponding *S*-functions on original scale. These are shown in Fig. 3 for all three components of particular matter, on a common relative scale from 0 to 100, for the sake of comparison. The results for PM_{10} and $PM_{2.5}$ are similar to those for PM_{10}–$PM_{2.5}$. The impact of salting on the concentration PM_{10} is almost as strong as on PM_{10}–$PM_{2.5}$, but more moderate on $PM_{2.5}$.

4.2. Results from the simplified models

In order to simplify the interpretation of the salting effects, we want to impose more structure on that part of model (1). Fig. 2 shows that it is reasonable to assume that the effect of salting decreases linearly (on log-scale) for about 5–15 days, after which it disappears. According to the figure, the duration may depend on the amount of magnesium chloride. However, it is difficult to estimate it precisely. Furthermore, it is hard to explain why the effect of salting should last longer using 20 g m^{-2} than the double amount. Hence, we will assume that the duration of the salting effect is the same for both amounts of magnesium chloride and all three pollution components. This corresponds to the simplified model (3). Our next step is to follow the estimation procedure, described in Section 3.2, in particular to assess the duration of the effect.

The left panel of Fig. 4 shows the total log-likelihood (Eq. (3)) as a function of the duration of the effect, θ^{salting}, for values between 1 and 20 days. Values outside this interval are considered to be implausible, and are not investigated. The best-fitting value of θ^{salting} is nine days, but the log-likelihood is rather flat between 5 and 20 days, indicating that the uncertainty may be large.

The β-coefficients are estimated for θ^{salting} fixed at nine, and the effects on original scale are calculated from (6). The explained variance (R^2) for the three models is about 70% (middle column of Table 3). The uncertainty in the magnitudes and

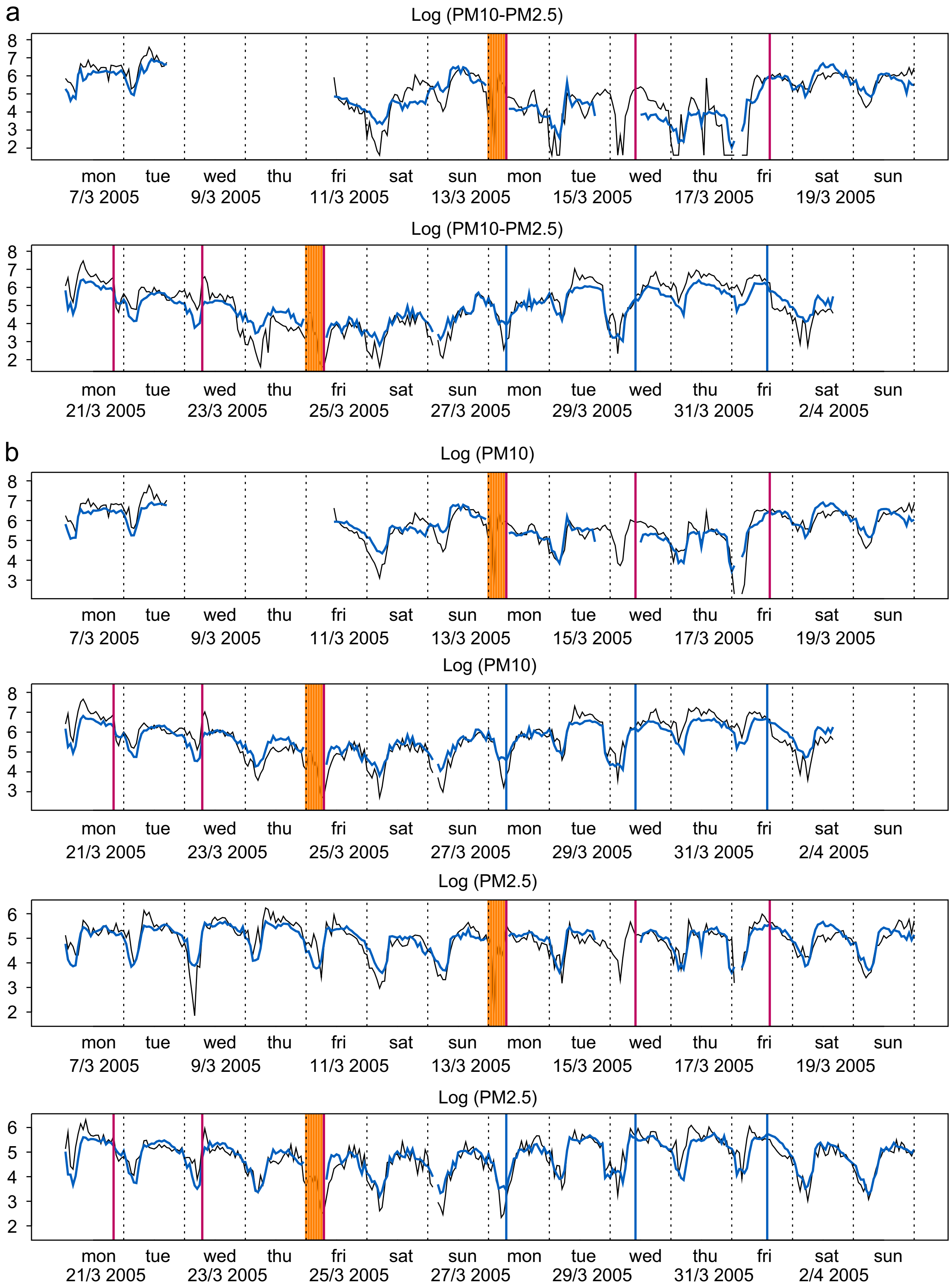

Fig. 5. Hourly values of the natural logarithm of (a) PM_{10}–$PM_{2.5}$, (b) PM_{10}, and $PM_{2.5}$ (black) together with fitted values from model (5) (blue) during the weeks 10–13 2005. Vertical lines have the same meaning as in Fig. 1.

duration of the effects is estimated with the bootstrap procedure described in Appendix A. This takes the residual autocorrelation and the uncertainty in the duration θ^{salting} into account. The estimates and their corresponding 95% confidence intervals are given in Table 2. As mentioned above, the duration of the effect is rather uncertain, with a confidence interval from 3 to 17 days. The estimated effect of salting is largest on the concentration of coarse particles PM_{10}–$PM_{2.5}$, close to 70% for both amounts of magnesium chloride. The impact on the level of PM_{10} is somewhat smaller. The estimates are 63% and 48% for amounts 20 and $40\,\mathrm{g\,m^{-2}}$, respectively. As for the concentration of fine particles, $PM_{2.5}$, the estimated effects are clearly smaller, namely 29% and 8%, and not even significant for the highest amount of magnesium chloride.

The effect of the two amounts of magnesium chloride on PM_{10}–$PM_{2.5}$ is similar. However, salting with $20\,\mathrm{g\,m^{-2}}$ is significantly more effective on PM_{10} and $PM_{2.5}$ than with the double amount. We cannot exclude the possibility that salting with $20\,\mathrm{g\,m^{-2}}$ is more advantageous for the air quality than the double amount, as magnesium chloride may be a dust source itself. Using too much salt might generate particle matter. However, the results may originate from artefacts in the data. More specifically, the amount $20\,\mathrm{g\,m^{-2}}$ was mainly used in the first half of the data period, and the double in the second half.

Therefore, we find it reasonable to estimate a common salting effect, as defined in model (5). Fig. 5 shows measured levels of the natural logarithm of PM_{10}–$PM_{2.5}$, PM_{10}, and $PM_{2.5}$ along with the corresponding fitted values from the model, indicating a reasonably good fit. Table 3 shows that the explained variance is almost as high as in model (3) with separate effects. As mentioned before, the residuals are autocorrelated (Fig. 6), which is accounted for when the uncertainty is calculated. Estimates of the duration and magnitude of the common effects are presented in Table 4. The duration is estimated to 10 days (see also the right panel of 4), with a confidence interval from 3 to 16. Applying magnesium chloride clearly binds coarse particles PM_{10}–$PM_{2.5}$ most effectively (70% reduction, confidence interval between 46% and 88%), whereas the level of fine particle is reduced only by 17% (confidence interval between 1% and 32%). The effect on PM_{10} is naturally somewhere in between these two (56%).

Table 3
Explained variance R^2 (in %), based on model (3), with separate effects of salting, and model (5), with common effect

Pollution component	Separate effects	Common effect
PM_{10}–$PM_{2.5}$	72.8	72.2
PM_{10}	75.3	74.9
$PM_{2.5}$	68.2	67.8

Note that the three effect estimates are mutually consistent: In the data, the coarse fraction PM_{10}–$PM_{2.5}$ constitutes about two thirds of the PM_{10} particles. If we combine the effect estimates for PM_{10}–$PM_{2.5}$ and $PM_{2.5}$, weighting with the fraction they represent, we get $\frac{2}{3}\cdot 70\% + \frac{1}{3}\cdot 17\% = 52\%$, which is similar to the estimate of 56% on PM_{10} from the model.

Fig. 7 illustrates how the common effect of salting diminishes with the time since it was performed, together with confidence intervals. This indicates how often one should apply magnesium chloride.

4.3. Sensitivity analysis

Finally, we have investigated how sensitive the main results in the previous subsection are to the various choices we have made in the modelling. First, we have varied the number of degrees of freedom in the s-functions to the half, one and a half and twice the number used in the main analysis, which was four for all functions except two (see Section 3.1). The results are given in Table 5 and should be compared to those in Table 4. The estimates of the duration are definitively sensitive to the choice of the number of degrees of freedom, which once again illustrates their uncertainty. The estimates of the salting effects, however, are comparable to those in the main analysis.

In an attempt to reduce the uncertainty of the duration estimate, we have so far assumed that the duration is the same for all three pollution components, but this assumption is questionable. Therefore, we have reestimated model (5), with separate durations for each pollution component, maximising the log-likelihood for each component (Table 6). Table 5 shows that the duration estimates change considerably, whereas the effect estimates are comparable to those in Table 4, especially the confidence intervals.

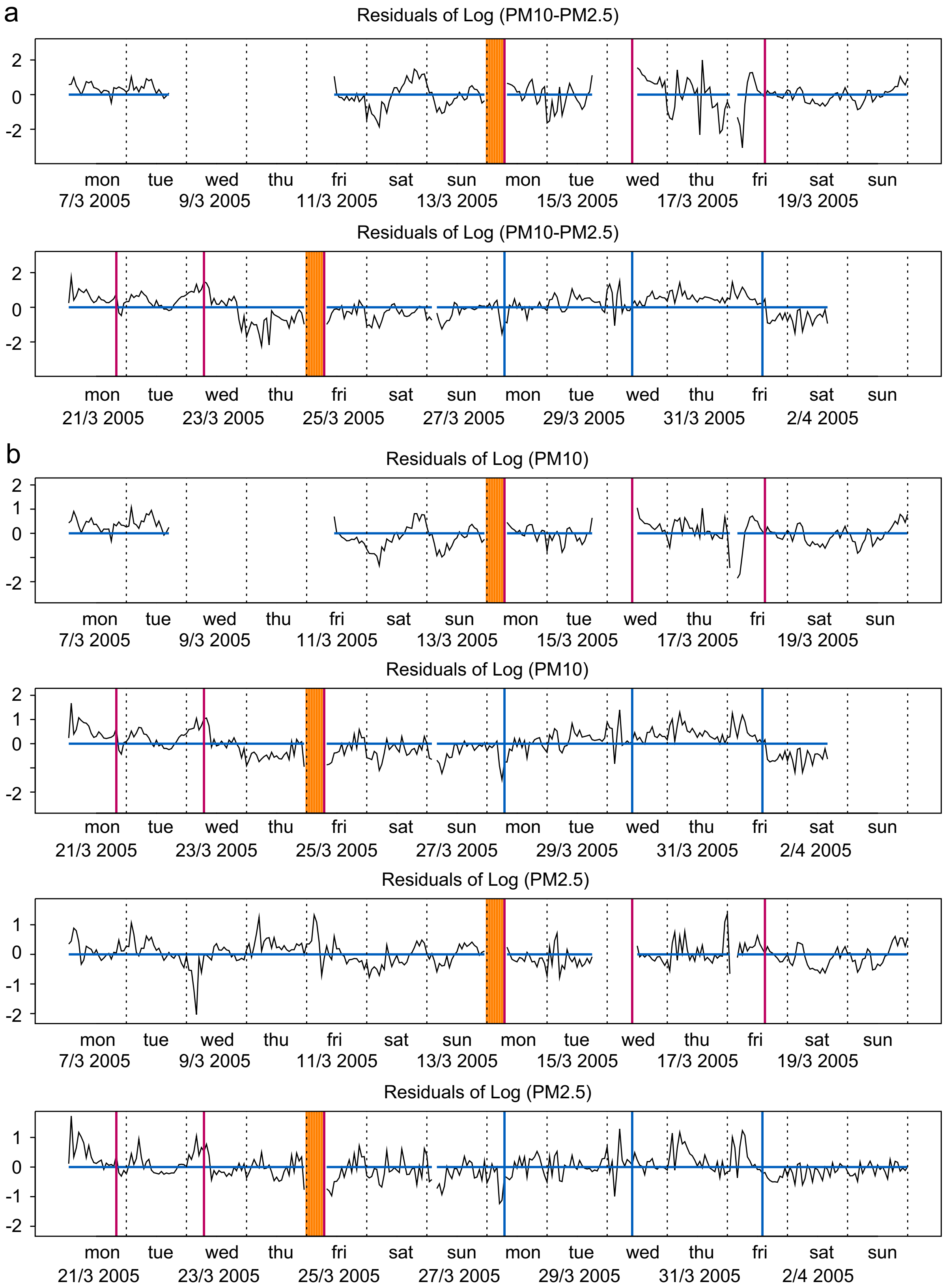

Fig. 6. Hourly values of residuals on logarithmic scale, corresponding to Fig. 5 (difference between the black and blue line in Fig. 5).

5. Discussion and conclusion

This work presents an analysis of the effect of salting with a magnesium chloride solution in a road tunnel on the concentration of particulate matter, more specifically PM_{10}, $PM_{2.5}$ and PM_{10}–$PM_{2.5}$. The model used for this purpose is a GAM on log-scale, having traffic counts, different meteorological conditions, sweeping, washing and salting as predictor variables. The model was simplified by linearising the effects of salting (on log-scale) in order to facilitate interpretation. The effect of

Table 4
Estimates and confidence intervals of the duration (in days) and magnitude (in %) of the immediate common effect of salting, based on model (5)

Pollution component	Duration of the effect		Common effect	
	Est.	95% conf. int.	Est.	95% conf. int.
PM_{10}–$PM_{2.5}$	10	(3, 16)	70	(46, 88)
PM_{10}	10	(3, 16)	56	(32, 72)
$PM_{2.5}$	10	(3, 16)	17	(1, 32)

Table 6
Estimates and confidence intervals of the duration (days) and magnitude (in %) of the immediate common effect of salting, based on model (5), with separate duration for each pollution component

Pollution component	Duration of the effect		Common effect	
	Est.	95% conf. int.	Est.	95% conf. int.
PM_{10}–$PM_{2.5}$	5	(3, 16)	53	(42, 83)
PM_{10}	16	(3, 20)	64	(32, 75)
$PM_{2.5}$	20	(1, 20)	28	(1, 43)

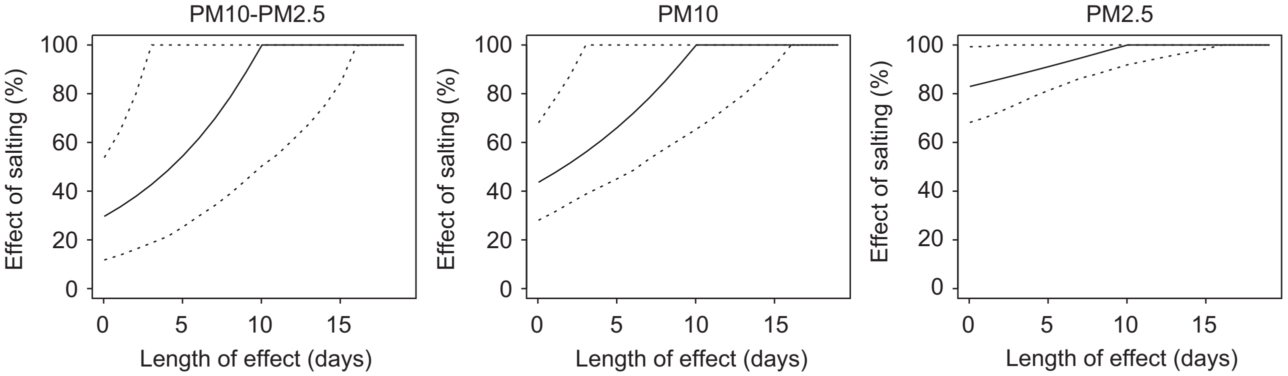

Fig. 7. Common effect of salting as a function of the time since it was carried out, with 95% confidence intervals.

Table 5
Estimates and confidence intervals of the duration (in days) and magnitude (in %) of the immediate common effect of salting, based on model (5), for various numbers of degrees of freedom in the *s*-functions (compared to the one used in the main analysis)

Pollution component	Duration of the effect		Common effect		Change in degrees of freedom
	Est.	95% conf. int.	Est.	95% conf. int.	
PM_{10}–$PM_{2.5}$	5	(3, 8)	56	(48, 78)	Half
PM_{10}	5	(3, 8)	44	(36, 62)	Half
$PM_{2.5}$	5	(3, 8)	12	(6, 26)	Half
PM_{10}–$PM_{2.5}$	17	(6, 19)	83	(60, 94)	50% higher
PM_{10}	17	(6, 19)	68	(41, 78)	50% higher
$PM_{2.5}$	17	(6, 19)	26	(−7, 43)	50% higher
PM_{10}–$PM_{2.5}$	5	(4, 17)	49	(43, 88)	Double
PM_{10}	5	(4, 17)	37	(29, 69)	Double
$PM_{2.5}$	5	(4, 17)	7	(−4, 31)	Double

salting is modelled as constant throughout the study period, but in reality it may vary with conditions we have not accounted for. The estimated effects should therefore be interpreted as average effects.

There may be interaction between the different pollution reducing measures. For instance, one could imagine that salting combined with washing has a larger effect than salting alone. An additional analysis (not shown here) revealed no significant interactions. Hence, interactions have not been included in our model. They could, however, be included in the model within the same framework.

The analysis revealed no clear effect from sweeping and washing. One reason may be that the number of times they were carried out is too low to reveal their effects, given the natural high variation in the PM levels. Another reason may be that the equipment used mainly removes particles coarser than PM_{10}. Further, sweeping and washing were always performed during the same period at night, in counter phase with the rush time. This may weaken their effect. However, our results agree with Norman and Johansson (2006) (and with references therein), who found no effect of sweeping and only a very small (6%) effect of washing.

The impact of salting on the concentration of particulate matter is clearly propitious. As one would expect, it is largest immediately after salting, and diminishes steadily afterwards. The concentration of coarse particles PM_{10}–$PM_{2.5}$ is the one most affected. The estimated effect immediately after salting is a 70% reduction compared to the corresponding PM level under the same conditions, but without salting. The impact on the level of PM_{10} is a little smaller (56%). The effect on the finest particles is modest (17%) and barely significant. Moreover, it appears to be more efficient to repeat the salt treatment more often than increasing the amount of binding material.

The duration of the salting effect is estimated to 10 days, but with a wide confidence interval from 3 to 16. This large uncertainty is due to the experimental design. Mostly, magnesium chloride was applied with short intervals. Hence, the PM levels immediately before salting are already reduced by a previous salting event. If the actions were more spread during the study period, the estimation uncertainty may have been reduced.

We conclude that the application of magnesium chloride solution is a potentially useful measure to reduce the level of particulate matter in a road tunnel. However, we have not considered possible adverse effects, such as potential damage to the environment or corrosion on vehicles and concrete constructions. In practice, before deciding to use magnesium chloride, one must consider the benefit from binding particulate matter versus the direct cost compared to alternative measures and possible adverse effects.

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Appendix A. Uncertainty assessment by the bootstrap

As mentioned in Section 3.3, the uncertainty of the parameters of interest (the immediate effect and the duration of the effect) is found by the bootstrap (Efron and Tibshirani, 1993). First, 1000 new bootstrap data sets are generated (reduced to 100 in the sensitivity analysis of Section 4.3). For each bootstrap data set, the model, including the duration of the salting effect, is re-estimated, yielding bootstrap estimates of the parameters of interest. Confidence intervals are calculated from these bootstrap replicates.

The bootstrap data sets are generated by resampling the residuals, keeping the predictor variables and the pattern of missing observations fixed. First, the empirical residuals are calculated by $\hat{\varepsilon}_t = z_t - \hat{z}_t$, where $z_t = \log(y_t)$ and $\hat{z}_t$ are the fitted values from the model estimated on the original data set. These are resampled, yielding bootstrap residuals ε_t^*, and corresponding bootstrap values of the response $z_t^* = \hat{z}_t + e_t^*$.

Ordinary bootstrap of the residuals, based on independent sampling with replacement, assumes no autocorrelation in the residuals, hence the uncertainty would be underestimated. Instead we use the stationary bootstrap (Politis and Romano, 1994; Lahiri, 2003), a variant of the block bootstrap (Carlstein, 1986; Künsch, 1989). Instead of resampling single residuals, blocks of consecutive residuals are drawn (with replacement) and joined together. The autocorrelation structure is preserved within each block, but not between blocks. If the block lengths are large enough, the breaks are less important. The block lengths are random, which avoids problems with periodicities (daily and weekly) in the data. Furthermore, the blocks are overlapping, and to avoid undesired edge effects, the end of the residual series are joined with the start (circular bootstrap).

We have implemented a modified version of the stationary bootstrap described above, taking into account special features of our data. Firstly, missing observations are handled specifically. When a potential block is drawn randomly from the empirical residuals, the block is rejected if its first residual is missing. If an accepted block contains missing residuals, only the first sub-series of observed residuals is used, i.e., only observed residuals are drawn and joined together. Finally, the original pattern of missing observation is imposed upon the bootstrapped residual series. Furthermore, the residual distribution may vary systematically with hour and day. Therefore, when a new block of residuals is drawn, it is drawn among blocks that start with the hour of week that fits with the end of the previous block.

In our case, the block lengths are uniformly distributed between two and a half and three and a half days, i.e. an average block length of three days. This is chosen by experimenting with average block length from 1 h (corresponding to independence) to several days. Too short block lengths result in an underestimation of the uncertainty. When the average block length is one day or more, the estimated uncertainties are stabilised.

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