Appendix: A proposed new statistical method for analysing the trends in measured concentration data at EMEP monitoring sites

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A1. Introduction

The EMEP measurement network provides data on air and rainfall concentrations for over 20 years at many sites across Europe, and is a valuable resource in seeking to assess the effects of reductions in pollutant emissions over the period. These monitoring data are available from the web database http://www.nilu.no/projects/ccc/emepdata.html. Barrett et al (2000) presented an analysis of trends relying primarily on the non-parametric Seasonal Kendall Test along with Sen's slope estimator. This was a very useful analysis, but it was clear from the figures in the report that there was no simple pattern of trend in either emissions or measured concentrations over the two decades. Also since changes in weather patterns will result in changes in observed concentrations with a stable emissions scenario, procedures that can adjust for meteorology before associating any decrease in concentration with emission control policies are useful. The purpose of this study was to investigate the use of modern statistical modelling procedures to see what advantages they could bring to the analysis of trend at one or more typical EMEP monitoring sites.

The statistical method proposed for this trend analysis is non-parametric general additive modelling, and it has several theoretical advantages:

1. the flexible trend line closely follows the data;
2. changes in pattern of trend over time are clearly presented;
3. the analysis can explicitly include the serial correlation between measurements at successive time points;
4. extra information (such as meteorological data) can be included as covariates;
5. the importance or influence of the covariates can be assessed statistically;
6. an extension of the analysis would allow regions with similar pollution trends to be objectively defined.

One part of the work was to see whether these theoretical advantages translated into a real improvement in understanding for this particular EMEP data set. The basis of the statistical model is that the fitted function at each point in time should be smooth and should be primarily influenced by data collected near that time point. Related applications include fitting moving-averages or spline functions to data collected at regular time intervals. These non-parametric methods contrast with techniques like regression where the aim is to find the best fit through all the data points of a single parametric function chosen in advance, such as a line or a quadratic curve.

One important issue when using smoothing procedures is to identify if there are sudden changes in the level of the data series. An analogy for the problem is walking along a rough track and then coming to a cliff edge. The assumption that the cliff is very like the last kilometre of track (relatively smooth) and so you can continue walking over the edge is rather foolish. As there were many cases in the EMEP monitoring data of both breaks in the recorded data series and of apparent changes in concentration, the first part of the study concentrated on developing a procedure to detect important discontinuities.

A2. Basic ideas for detecting discontinuities

A discontinuity in these EMEP concentration series can be defined as a point with an abrupt change in the mean level of the pollutant concentration, where the change can be temporary or permanent. The objective was to determine, solely from the data, the time points where the data series may be discontinuous. The procedure was developed from a diagnostic for flagging discontinuities in one-dimensional non-parametric regression (Bowman and Pope, *pers. com.*). The idea is to compare, at each point in the data series, two linear smoothes of the data. One smooth uses only data to the left of the test point and the other uses only data to the right of the test point. If there is a discontinuity, then the left smooth will predict a significantly different value at the test point from the value predicted by the right smooth.

The smoothing procedure chosen was local linear regression with weights, taken from a Normal density function, which decrease as the distances between time points increase. This is a very flexible smoother and was able to represent the observed data series well. There is positive serial correlation in the data, so successive data values are more similar than implied by the usual random error structure for the residuals. The estimates of variance, and hence the test statistic for assessing the difference between left and right smoothes, were adjusted to remove the effect of this correlation.

A3. Finding discontinuities in practice

Tests for discontinuities were applied to the concentrations of SO_2, SO_4 in air and SO_4 in precipitation for most EMEP monitoring stations. Recorded concentrations are generally daily mean values with considerable day-to-day variation and a skew frequency distribution. The daily variability was to great to detect important discontinuities at that time resolution, but moving to a weekly measure of concentration gives reduced variation on the weekly timescale and no loss of effective time resolution for locating a discontinuity over the 20 year period. Preliminary analysis of the daily data also detected two kinds of repeated patterns in the series, 'days within week' and 'weeks within year'. Both these effects were modelled and removed before calculating *the weekly means of the logarithm of the seasonally adjusted daily data*, which are the data used for the subsequent discontinuity detection process.

Figure A1 illustrates the procedure for SO_2 concentrations at Vreedepeel (NL10). Because of the edge bias associated with any smoothing procedure, fifty data points (or approximately 1 year of data) at the start and at the end of the series were excluded from the tests for discontinuity, although these data points were used in estimating the smooth functions. The plot of the left and the right smoothes are shown along with a shaded region indicating the confidence band for the difference between the smooth functions, giving a fundamental guide

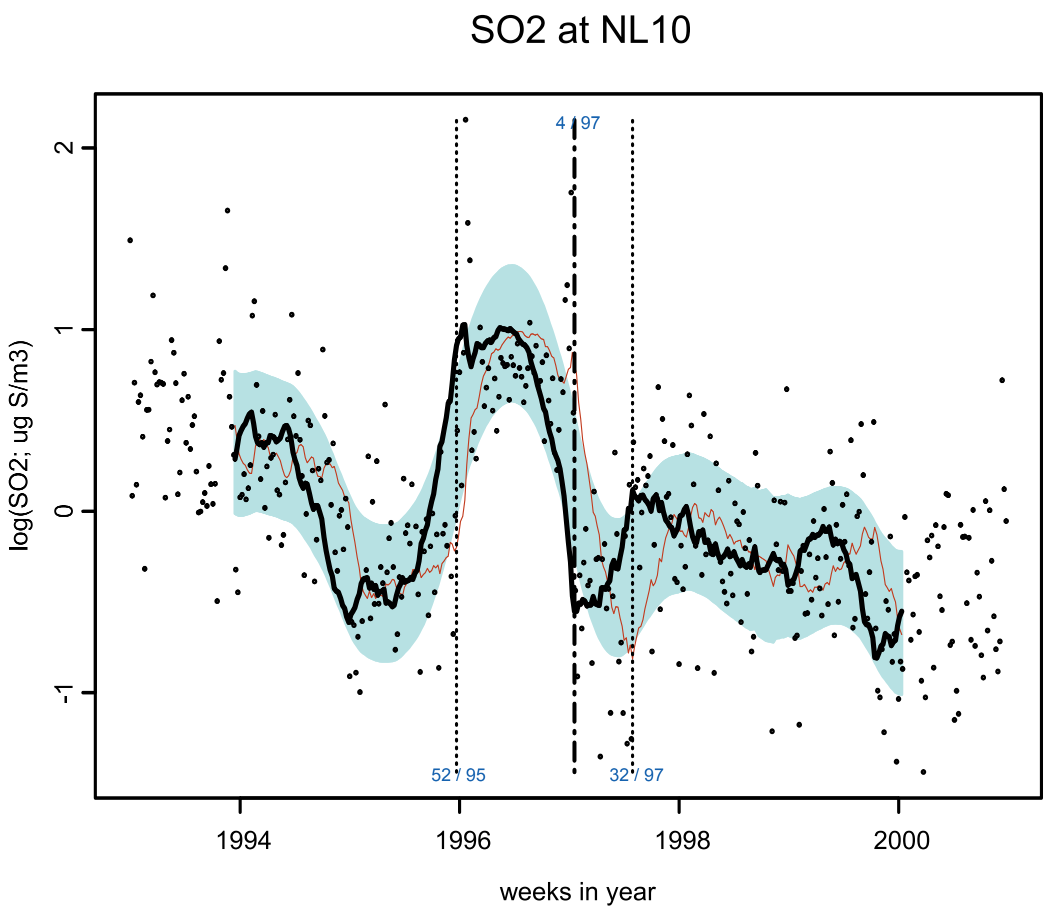

Figure A1: Left and right smoothes for SO_2 at Vreedepeel (NL10) showing the 99.75% (or 1 in 400) confidence band and the time points where three discontinuities were detected

to places where discontinuities may be found. If both left and right smoothes leave the shaded region at the same point then this suggests the possible presence of a discontinuity. The vertical lines on Figure A1 indicate where the discontinuities were detected, with the bold line marking the most significant discontinuity. It is clear from the plot that the vertical lines do not identify a precise week when a discontinuity occurs, as, for example, occurs when the left and right smoothes are outside the confidence band on week 52 in 1995 and for four subsequent weeks. The significance level for the confidence band was set empirically at 1 in 400, as this level appeared to identify the major discontinuities while screening out the many minor changes in mean concentration.

Two further outputs help with the interpretation of the discontinuity. Figure A2 shows the smoothed trend in the different segments of the time series and Table 1 provides the list of dates when left and the right smoothes are significantly different. As the analysis is on a logarithmic scale, the jump (Table 1) between the two smoothes gives the $\log_e$ of the ratio of

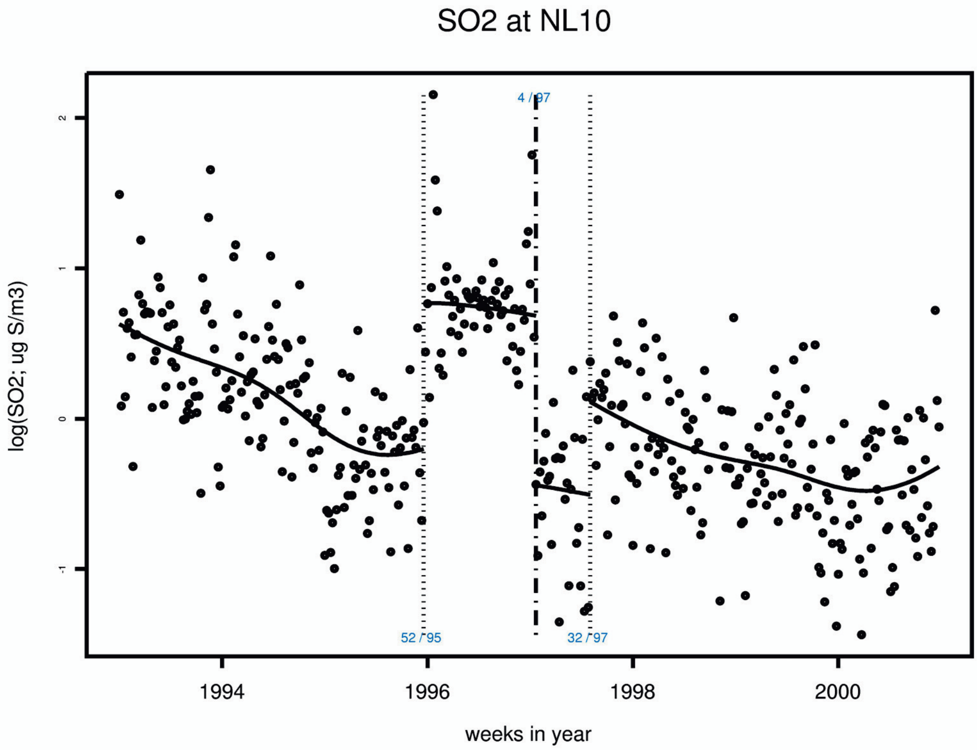

Figure A2: Smoothed trends between the three detected discontinuities for SO_2 at Vreedepeel (NL10)

the mean weekly level from the right smooth to the mean weekly level from the left smooth. The SO_2 concentration dropped from 1993 to 1995 and then there was a jump so that the mean concentration in 1996 was approximately 3 times larger than in 1995. In the first part of 1997 the concentration dropped to 0.75 of the 1995 level and subsequently increased in summer 1996 to 1.75 times the 1995 level before continuing its longer-term decline.

The SO_2 concentrations at another site in the Netherlands, Kollumerwaard (NL09), are plotted in Figure A3 and data on the discontinuities detected reported in Table A1. The idea of 'common national discontinuities' can be explored by looking for the possible presence of discontinuities across the stations in a country. For example, similar discontinuities for SO_2 at Vreedepeel and Kollumerwaard occur to give elevated concentrations at both sites in 1996 compared to 1995 - for Kollumerwaard the 1996 concentration was 6 times the 1995 level but it returned in 1997 to 1.3 times the 1995 level. There were also elevated concentrations of SO_4 in air at Kollumerwaard in late 1995 and early 1996 but this was not reflected at Vreedepeel.

Discontinuities can be attributed to several reasons such as changes in emissions, changes in laboratory procedures, use of new instrumentation, or occurrence of particular climatic conditions. With a temporary change in laboratory analysis shown as two discontinuities, the

Table A1: Discontinuities detected at Vreedepeel (NL10) and Kollumerwaard (NL09)

Site	Compound	Week	Year	Jump
NL10	SO2	52	1995	1.111
NL10	SO2	4	1997	-1.402
NL10	SO2	32	1997	0.928
NL10	SO4air	6	1997	-0.855
NL09	SO2	52	1995	1.850
NL09	SO2	53	1996	-1.561
NL09	SO4air	40	1995	0.671
NL09	SO4air	17	1996	-0.626
NL09	SO4prec.	15	1994	1.012
NL09	SO4prec.	53	1994	0.995

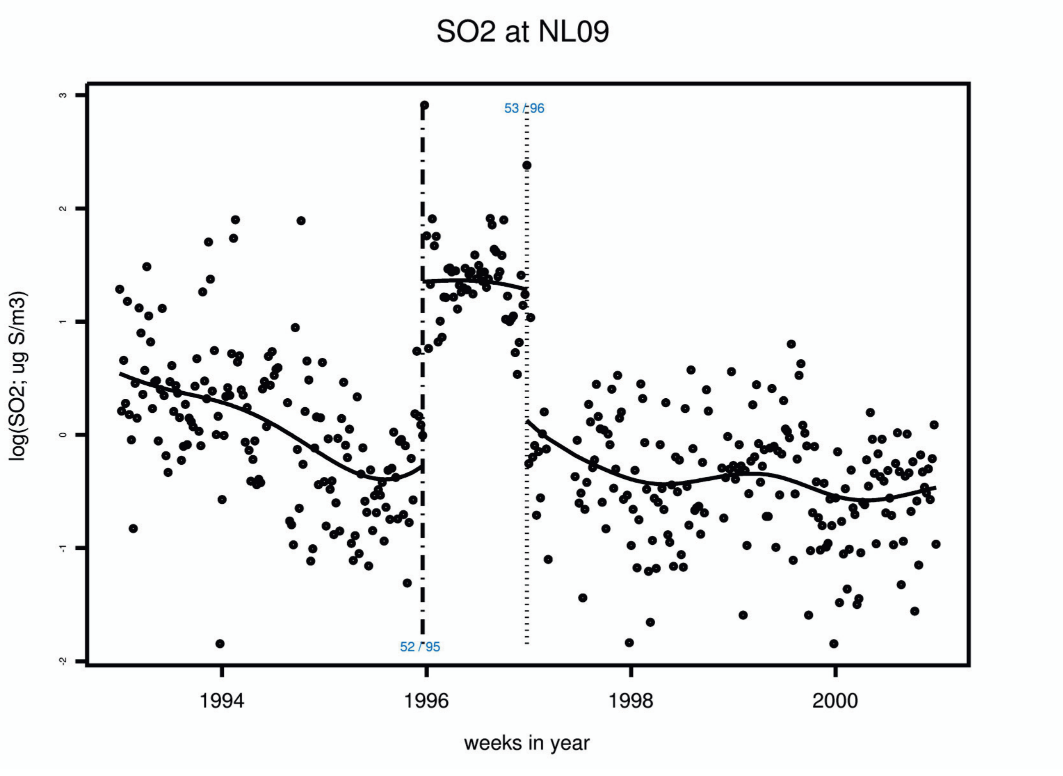

Figure A3: Smoothed trends between the two detected discontinuities for SO_2 at Kollumerwaard (NL09)

trend could be modelled with some adjustment for the period between the discontinuities. A discontinuity related to a change in emissions could be confirmation that a policy has succeeded without any further trend analysis, or it might be more sensible to split the data at the discontinuity and model the trend in two parts. A change in climatic conditions resulting in a discontinuity may suggest the inclusion of covariates in the trend model. The statistical process on its own does not determine what is the best action to take, if any, following detection of a discontinuity.

The discontinuity tests were applied to 113 sites across 16 European countries covering the period 1977-2000. Possible discontinuities were detected across countries and across compounds, revealing some interesting features of the data, and this test appears to be useful both as an enhanced screening of the data series and to identify times when important changes may have occurred. In all cases, the purpose of this study was to identify possible discontinuities and report these to the data providers so that they could (a) determine if there was a scientific reason for the discontinuity and (b) decide whether or not there should be a consequent adjustment to the stored data series.

A4. Modelling Trends

From the discontinuity study it was clear that trends at many sites were neither monotonic nor reasonably approximated by a linear measure, assumptions implicit in the use of the non-parametric seasonal Kendall test and the Sen slope estimator in the overall analysis of trends (Barrett *et al*, 2000). It was also clear from many discussions that scientists saw the relationship between trends and changes in meteorology as a very important factor in understanding the atmospheric processes. The aim of this second part of the study was to adapt non-parametric general additive modelling procedures and apply them to the EMEP measurement network data, so providing smoothed trends adjusted for meteorological effects.

The availability of meteorological data was a major issue for this study and there were only seven sites with both long time series and meteorological data available for the analysis: Eskdalemuir (GB02), Westerland/Wenningstedt (DE01), Lagenbrügge/Waldhof (DE02), Schauinsland (DE03), Deuselbach (DE04), Brotjacklreigel (DE05) and Kosetice (CZ03). The daily data were adjusted to remove the repeating pattern 'days within week' and then *the weekly means of the logarithm of the adjusted daily data* were calculated - note that the seasonal pattern was not removed in contrast to the data used for the discontinuity detection. Use of the weekly values gives a more tractable computational challenge without losing information important to long term trend analysis.

The basic model fits the response variable, here a concentration, to the sum of a set of arbitrary functions of the selected covariates. The covariates considered were *time, seasonal effects, rainfall, temperature, humidity and a composite wind vector* (an averaged wind speed and wind direction). The procedure finds smooth functions of each covariate representing the effect of that individual covariate on the response, and puts these covariate functions collectively into an additive framework to best predict the observed response data. An advantage of the additive approach is that, after the fitting, it is possible to plot the individual covariate functions separately to examine their role in modelling the response. A backfitting algorithm was used with kernel smoothers, a generalisation of the local linear smoothers used in the discontinuity analyses, and the procedure accounted for serial correlation in the responses. Circular smoothers were seasonality and the wind vector, so that, for example, fitted for seasonality and the wind vector, so that, for example, when fitting a function of weeks of the year the effect of week 53 was close to the effect of week 1.

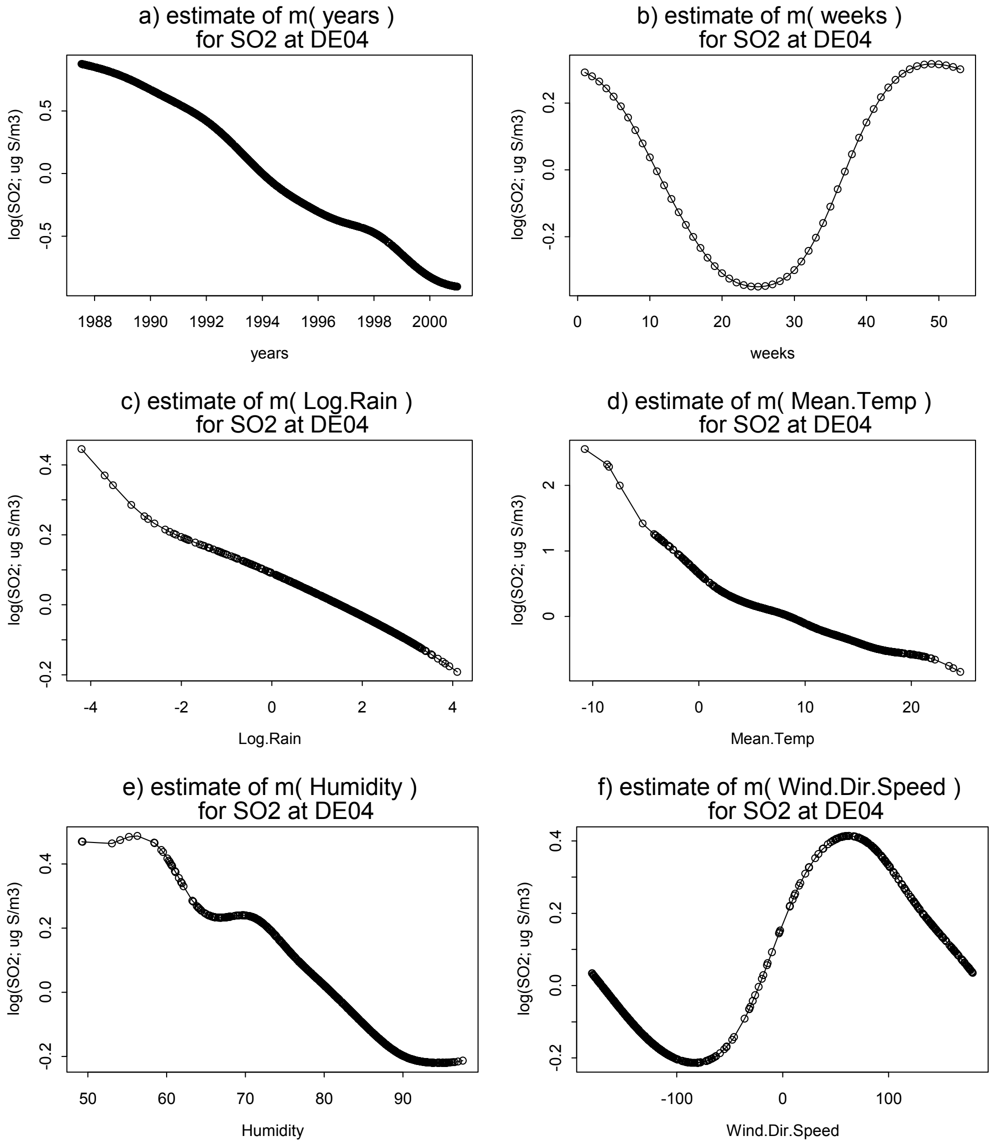

Figure A4: Response of SO_2 concentration to covariates at Deuselbach (DE04): (a) year, (b) week within year, (c) precipitation, (d) temperature, (e) relative humidity, and (f) wind vector.

The effects of the various covariates are illustrated for Deuselbach (DE04) in Figure A4. The components indicate that higher SO_2 concentrations (over the year) are associated with the winter period. Increased concentrations are associated with mean temperatures well below zero and, as expected, both rainfall and high humidity decrease the concentration. Higher SO_2 concentrations are associated with wind directions towards the east. The covariate effect of year is probably most interesting, indicating that concentrations dropped from 1988 to 1996 and then continued decreasing, but at a lower rate.

Plots of trends with time corrected for both seasonal effects and for meteorology are shown for the seven sites in Figure A5. The inclusion of the meteorological covariates was statistically significant within the model and had the effect of bringing the fitted values much closer to the data - without the meteorology there was seasonally variation around the blue trend line but the fitted values captured less than half of the variability associated with the observations. While the meteorology improved significantly the ability of the model to capture the patterns in the data, the underlying trend (the blue lines in Figure 5) are not much affected by the inclusion or exclusion of the meteorological variables.

A5. Conclusions

The non-parametric general additive modelling procedures were successfully applied to the EMEP air and rainfall concentrations of sulphur compounds at many sites across Europe, The results confirmed that flexible trend lines could be fitted which do closely follow the data and these trends can be presented so that the changes in pattern of trend over time is clear. As expected there was serial correlation in the data but this was successfully modelled. Discontinuities in the data series are a major problem and a procedure for detecting discontinuities was developed and hopefully will be applied in future data screening procedures.

The statistical model allowed the inclusion of covariates and it was found the meteorological information was necessary to model the variability in weekly averaged concentrations (on a logarithmic scale), both being statistically significant and also changing the model fitted values considerably – a clear visible improvement. The modelled trend was not substantially altered by the inclusion of meteorological covariates but there was a substantial change in the estimates of variance. This analysis also indicates that a substantial fraction of the variation in weekly averaged concentrations is related to meteorological effects.

Although meteorological covariates are necessary for an adequate statistical model of the trend, at sites where these are not available the annual trend pattern can be fitted using a model with only seasonal components. It is not possible at these sites to obtain an adequate estimate of the variance.

For any procedure to build up tests of differences between sites or to generate regional trend models, the meteorological data are a vital component of the analysis. It was found to be very difficult to obtain good site-based observed meteorology and this was a major factor in reducing the scope of this study. It is suggested that future studies should look at the possibility of using output from models rather than site measurements.

The comparison of the trends from the general additive model for the 7 sites are compared in Figure A6 with the trend lines given by the non-parametric Seasonal Kendall Test along with Sen's slope estimator as reported by Barrett *et al* (2000) - the red lines on the plots are Sen's slope estimators with their relevant uncertainty bands. While for some of the sites, the linear slope estimator and the smoothed trend are in reasonable agreement, for other sites there seems to be little evidence in the data to support the fitted Sen slope estimator.

In summary, there appeared to be no case to support the simple linear summary of trends at EMEP monitoring sites. Trends were not linear and smoothing procedures are available which capture and present the trends visually. Models are available to test the influence of covariates and it was found that meteorological data are necessary covariates. The models can be extended to establish spatial trend patterns and to include (and test) the effects of changes in pollutant emissions. Therefore there is a strong case for future trend analyses of EMEP monitoring data to use more appropriate statistical models.

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Reference

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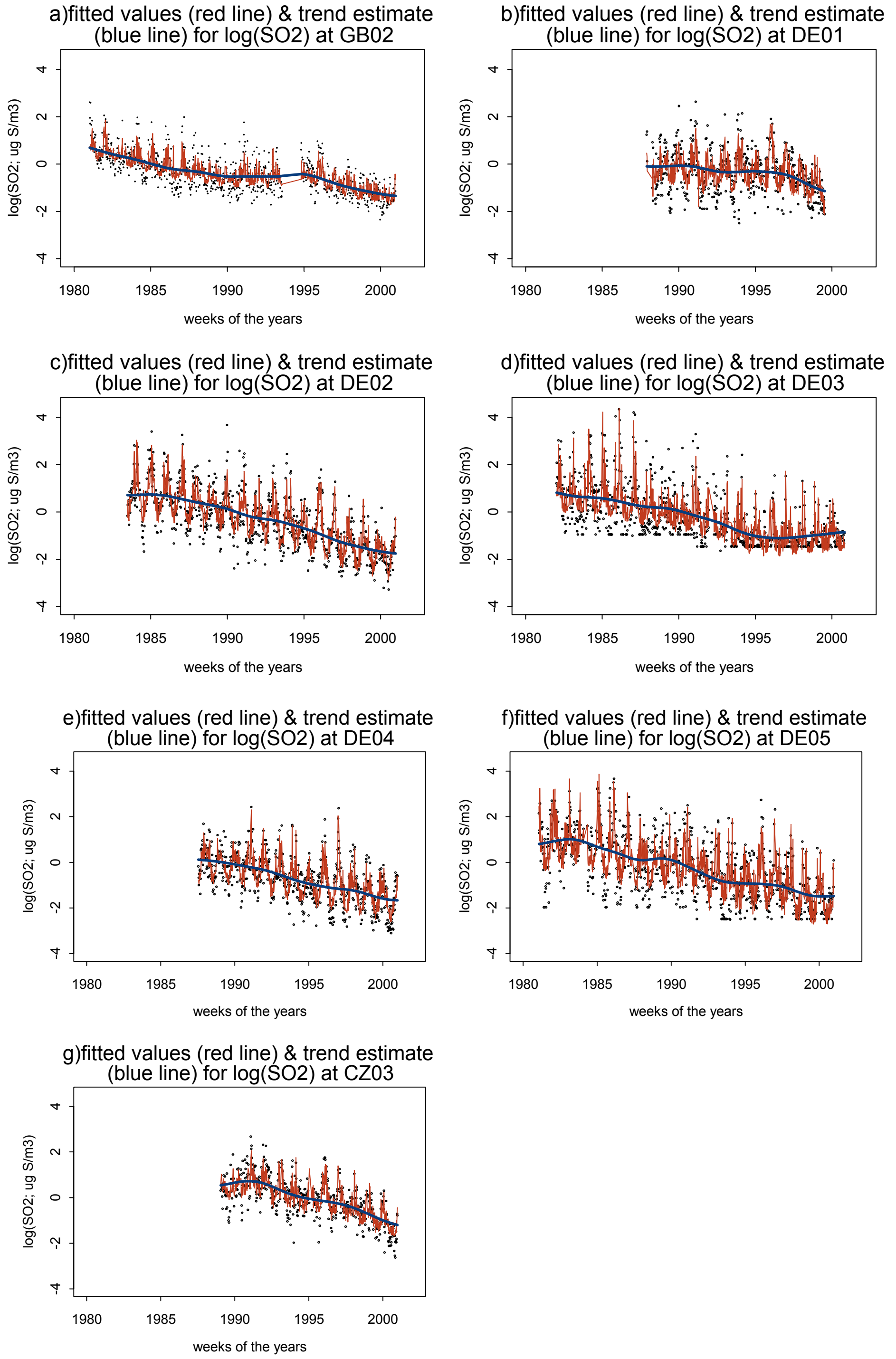

Figure A5: Fitting the model (red line) to logarithm of SO_2, with trends (blue lines) corrected for seasonality and meteorology at:
(a) Eskdalemuir (GB02), (b) Westerland/Wenningstedt (DE01),
(c) Lagenbrügge/Waldhof (DE02), (d) Schauinsland (DE03),
(e) Deuselbach (DE04), (f) Brotjacklreigel (DE05),
(g) Kosetice (CZ03).

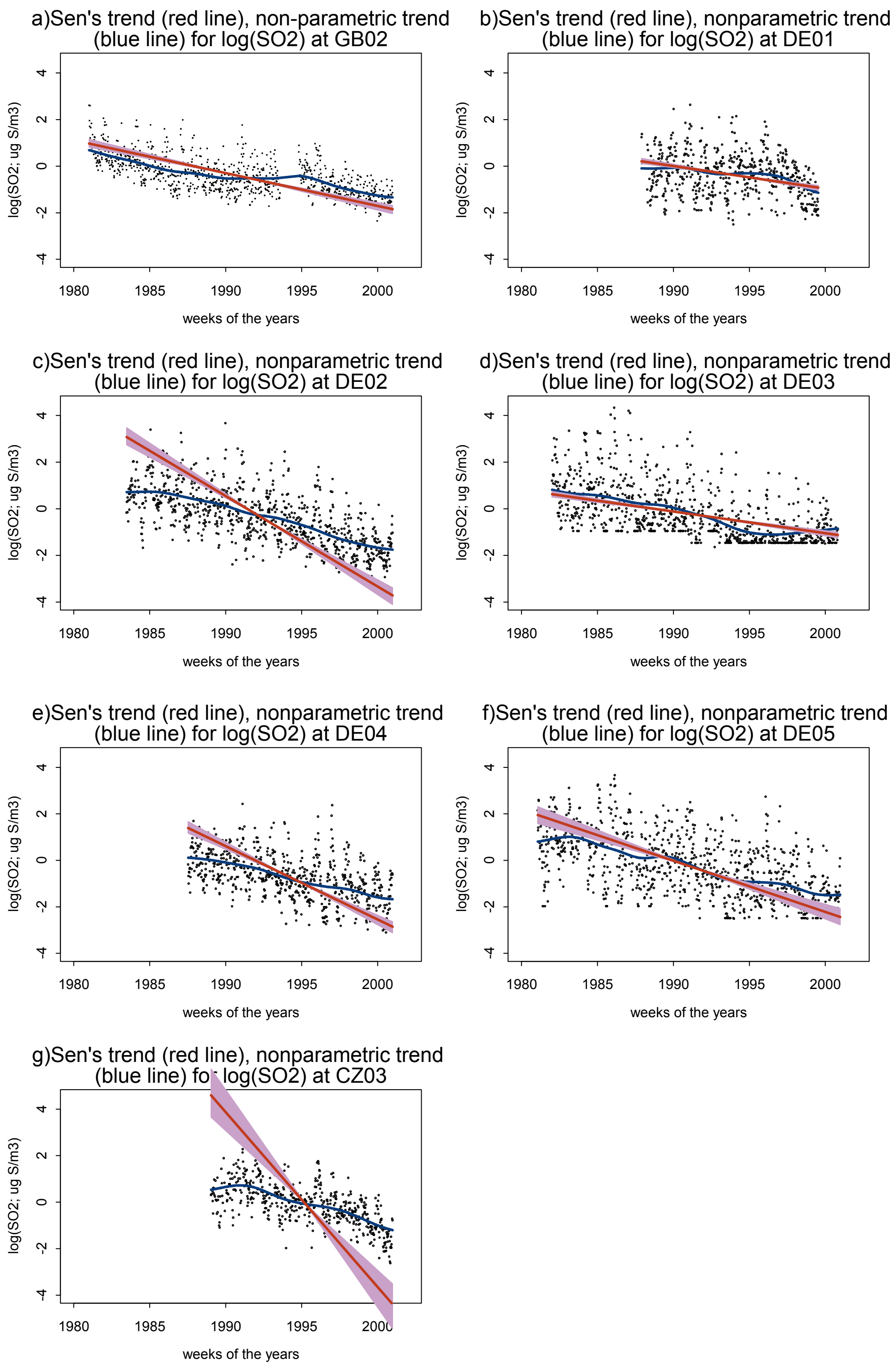

Figure A6: Sen's trend estimate (red line) and non parametric trend estimates (blue line) for $log_e(SO_2)$ at: (a) Eskdalemuir (GB02), (b)Westerland/Wenningstedt (DE01), (c)Lagenbrügge/Waldhof (DE02), (d)Schauinsland (DE03), (e)Deuselbach (DE04), (f) Brotjacklreigel (DE05), (g)Kosetice (CZ03).