

**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

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# Robust extraction of baseline signal of atmospheric trace species using local regression

**A. F. Ruckstuhl<sup>1</sup>, S. Henne<sup>2</sup>, S. Reimann<sup>2</sup>, M. Steinbacher<sup>2</sup>, B. Buchmann<sup>2</sup>, and C. Hueglin<sup>2</sup>**

<sup>1</sup>Institute for Data Analysis and Process Design, Zurich University of Applied Sciences, Winterthur, Switzerland

<sup>2</sup>Empa, Swiss Federal Laboratories for Materials Science and Technology, Laboratory for Air Pollution and Environmental Technology, Dübendorf, Switzerland

Received: 13 October 2010 – Accepted: 18 November 2010 – Published: 7 December 2010

Correspondence to: Christoph Hueglin (christoph.hueglin@empa.ch)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

The identification of atmospheric trace species measurements that are representative of well-mixed background air masses is required for monitoring atmospheric composition change at background sites. We present a statistical method based on robust local regression that is well suited for the selection of background measurements and the estimation of associated baseline curves. The bootstrap technique is applied to calculate the uncertainty in the resulting baseline curve. The non-parametric nature of the proposed approach makes it more flexible than other commonly used statistical data filtering methods. Application to carbon monoxide (CO) measured from 1996 to 2009 at the high alpine site Jungfrauoch (Switzerland, 3580 m a.s.l.) demonstrates the feasibility and usefulness of the proposed approach. The determined average annual change for the 1996 to 2009 period as estimated from filtered annual mean CO concentrations is  $-2.1 \pm 1.3$  ppb/yr. For comparison, the linear trend of unfiltered CO measurements at Jungfrauoch for this time period is  $-2.9 \pm 1.5$  ppb/yr.

## 1 Introduction

Background monitoring sites are the locations for observing the composition of the clean and remote atmosphere and for detection of long term changes and trends in important atmospheric trace species. However, many background monitoring sites are frequently affected by air masses that are influenced by local or regional sources or air masses that are representing certain atmospheric layers. Air samples taken at these locations are temporarily not representative of well-mixed background air. Hence, data filtering is often an essential part of the analysis of data from those sites. For example, data filtering was applied for trend estimations (Thoning et al., 1989; Novelli et al., 1998; Schuepbach et al., 2001; Novelli et al., 2003; Zellweger et al., 2009), for evaluation of source regions and corresponding emission estimates (Prinn et al., 2001; Cox et al.,

AMTD

3, 5589–5612, 2010

### Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2005; Reimann et al., 2005; Greally et al., 2007), as well as for modeling of long-range transport of trace gases (Ryall et al., 1998; Balzani Lööv et al., 2008).

5 Methods for identification of background measurements are often based on chemical parameters (trace gas concentrations or ratio of trace gases, e.g., Carpenter et al., 2000; Zanis et al., 2007) or take advantage of the knowledge on the transport processes of polluted air masses to the background site (meteorological filters). Meteorological filters have been applied in a number of studies utilizing data from the Swiss high-alpine site Jungfrauoch (JFJ), 3580 m a.s.l. (Forrer et al., 2000; Zellweger et al., 2003; Henne et al., 2005) for discrimination between disturbed and undisturbed free tropospheric air. In Zellweger et al. (2003), measurements that were identified as being influenced by föhn events, synoptical lifting, or thermally-induced vertical transport were excluded from further analysis. Another meteorological data filtering approach is the analysis of air mass origin as demonstrated for example by Derwent et al. (1998) and Balzani Lööv et al. (2008).

15 Statistical methods are an alternative to the application of chemical parameters and meteorological filters. In contrast to these approaches, statistical methods have not to be adapted to the conditions at individual measurement sites and can therefore be applied generally making background data of various stations easier to compare. Common statistical methods rely on the identification of measurements that deviate from a smooth curve fit to the data (Novelli et al., 1998; O'Doherty et al., 2001). For example, Novelli et al. (1998) fitted a second order polynomial plus the sum of four harmonics to daily carbon monoxide (CO) data from the NOAA/CMDL network and applied two low-pass filters to the model residuals. Measurements with large distance to the smoothed curve (defined as the sum of the parametric model fit and the smoothed residuals) were considered as outliers and flagged. The routine was then iteratively applied without the flagged measurements until no additional outliers were identified. In a subsequent study, a modified version of this method was applied (Novelli et al., 2003). In contrast to the earlier method, the model residuals were converted to the frequency domain with a Fourier transform algorithm and filtered by a low-pass and

---

**Extraction of background concentrations**

A. F. Ruckstuhl et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



a high-pass filter. The filtered residuals were then transformed back into the time domain and added to the fitted function resulting in the desired smooth curve.

Another statistical method for identification of background measurements was used in several studies from the Global Atmospheric Gases Experiment/Advanced Global Atmospheric Gases Experiment (GAGE/AGAGE), see e.g. Simmonds et al. (2001). This approach is based on a three step procedure and is described in detail by O'Doherty et al. (2001). In brief, the pollution events on a selected day are first identified by applying a second-order polynomial to the daily minima over the time period from 60 days before and 60 days after the selected day. The polynomial fit is then subtracted from the data and the variability  $\sigma$  of the residuals is estimated using only the data that are smaller than the median of the residual distribution. All measurements in the middle day of the 121 day period with residuals exceeding  $3\sigma$  are flagged as being "polluted". In a next step, the complete cycle of flagging data is repeated except that all data points that were marked in the previous cycle are excluded. At the end of this step, measurements between  $2\sigma$  and  $3\sigma$  above the median of the residuals are marked as "possibly polluted". In a final third step, all data points that are marked as "possibly polluted" are also labeled "polluted" if they are immediately adjacent to a polluted data point.

Meteorological filters and statistical approaches are frequently combined. For example, Thoning et al. (1989) applied and compared different selection methods based on daytime and short term variability of carbon dioxide ( $\text{CO}_2$ ) at Mauna Loa, Hawaii, to identify data that are influenced by local phenomena and not representative of well-mixed background air. Then, additional statistical filtering similar to the method used by Novelli et al. (2003) was done for removal of remaining short-term variability in the data.

In this study, a novel statistical approach for extracting background concentrations from measurements is presented. It is based on robust local regression (Cleveland, 1979) and is called REBS (robust extraction of baseline signal). It is a modified version of a technique that was called RBE and developed for baseline removal from chemical analytical spectra (Ruckstuhl et al., 2001). A difference to the RBE

**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



technique is the estimation procedure for the scale parameter  $\sigma$  of the measurement error. Here we either use only the negative residuals as it is similar to the method by O'Doherty et al. (2001), or preferably only the residuals below the mode of the residuals distribution are used. The precision of the measuring instrument can be considered as a lower bound for the estimate of the scale parameter.

In the next section, the REBS method will be introduced in detail. The proposed method can easily be applied to time series from any background site. This is demonstrated by applications to the long-term CO measurements from Jungfraujoch. The results are compared with those from a data filtering and baseline fitting technique that is similar to the approach of Novelli et al. (2003). The REBS algorithm can be found in the IDPmisc library (Ruckstuhl et al., 2010) for the statistical software package R (R Development Core Team, 2010) and can be downloaded from a CRAN server or received from the authors. Note that the current version of the REBS function in IDPmisc (rfbaseline) does not include the uncertainty estimation using the bootstrap method as described below.

## 2 Robust extraction of baseline signal

### 2.1 The REBS technique

In this section, we introduce a statistical approach for extracting background concentrations from trace gas measurements. The presented approach is a modified version of the robust baseline estimation (RBE) technique that was developed for baseline removal from chemical analytical spectra (Ruckstuhl et al., 2001).

We can consider the observed concentrations  $Y(t_i)$  to be defined by

$$Y(t_i) = g(t_i) + m(t_i) + E_i, \quad (1)$$

where  $g(t_i)$  is the background concentration and  $m(t_i)$  is the contribution of regionally polluted air masses at times  $t_i$  (called regional signal henceforth). The measurement

## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



errors  $E_j$  are assumed to be independent and Gaussian-distributed with mean 0 and variance  $\sigma^2$ . If the regional signal  $m(t_i)$  is zero in a time period around  $t_0$ , the background signal  $g(t_0)$  can be estimated even when the form of the curve  $g$  is unknown. If we can assume that  $g$  is smooth, then a method for estimating the curve  $g$  is to apply linear regression modeling locally. Hence the curve  $g(t_i)$  can be approximated as linear in a sufficiently small neighborhood around any given time point  $t_0$ . One can simply apply the least-squares technique to a fraction of the data around  $t_0$ , or alternatively, one can incorporate a weight scheme into the least squares problem that decreases the influence of data points in proportion to their distance from  $t_0$ . Such estimators are described, e.g., in Cleveland (1979), in Simonoff (1996) or in Fan and Gijbels (1996).

Separating the three components in Eq. (1) is an ill-posed problem without additional information. We argue here for assuming that the background signal  $g$  must vary very slowly relative to any contributions of regional signal and that this regional signal  $m$  is zero at many time points  $t_j$ . Then the basic idea of the “robust extraction of baseline signal (REBS)” technique is to regard measurement points  $Y(t_j)$  as outliers if  $m(t_j) \gg \sigma$  which is satisfied at time points  $t_j$  that show clear contributions of regionally polluted air masses. Since the regional signal must be non-negative, that is  $m(t_j) \geq 0$ , all of the outliers point in the same direction and thus we have an asymmetric contamination of the background signal. In such a case, Ruckstuhl et al. (2001) suggest estimating the background signal by applying their robust baseline estimation technique. That is, solve

$$\hat{\theta}(t_0) = \arg \min_{\theta} \sum_{i=1}^n w_r(t_i) K \left( \frac{t_i - t_0}{h} \right) \times [y_i - \{\theta_0 + \theta_1 (t_i - t_0)\}]^2. \quad (2)$$

Note that the resulting estimated parameters  $\hat{\theta} = (\hat{\theta}_0, \hat{\theta}_1)^T$  depend on  $t_0$ . Thus,  $\hat{\theta}_0(t_0)$  is an estimate of  $g(t)$  at  $t_0$  and is better named  $\hat{g}(t_0)$ . To obtain an estimate of the whole background signal  $g$ , we solve Eq. (2) for a set of time grid points  $t_0 = \tilde{t}_k, k = 1, \dots, K$  and interpolate them linearly (the original time points  $t_j$  may also be selected as time grid points  $\tilde{t}_k$ .)

## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



As kernel weight function  $K[(t_i - t_0)/h]$  the tricube kernel

$$K\left(\frac{t_i - t_0}{h}\right) = \left[ \max\left\{1 - \left|\frac{t_i - t_0}{h}\right|^3, 0\right\} \right]^3 \quad (3)$$

is used which descends smoothly to zero and is zero outside the neighbourhood  $t_0 \pm h$ . To down-weight the outlying regional signal  $m(t_i)$ , an asymmetric robustness weight  $w_r(t_i)$  is introduced:

$$w_r(x_i) = \begin{cases} 1 & \text{if } r_i < 0 \\ \left[ \max\left\{1 - (r_i/b)^2, 0\right\} \right]^2 & \text{otherwise,} \end{cases} \quad (4)$$

where  $r_i = [y_i - \hat{g}(x_i)]/\sigma$ . The standard choice for the tuning constant  $b$  is 3.5, however, any value for  $b$  within 3 and 4 seems appropriate. On one hand, outliers might receive too much weight when  $b$  is larger than 4. On the other hand, the smaller the tuning constant the higher the systematic error in time series with no or a very small number of polluted measurements. It should, however, be noted that the use of asymmetric robustness weights also helps to ensure that the fit converges to an acceptable solution.

A critical issue for the REBS technique is how wide the local neighborhood should be (i.e., what value of so-called bandwidth  $h$ ). A number of suggestions have been advanced for automatically determining an appropriate bandwidth from the data (Simonoff, 1996; Fan and Gijbels, 1996). However, these approaches would lead to reasonable bandwidths  $h$  for estimating the “total” signal  $g(t_i) + m(t_i)$ , which is not our goal. A more problem specific consideration is the following: if, in a local neighborhood of  $t_0$  consisting of  $d$  data points, at least  $d/2$  of them are seriously affected by the regional signal  $m$ , then the robust local regression estimator is more likely to estimate  $(g(t_0) + m(t_0))$  than  $g(t_0)$ . To avoid such a failure, we can require that  $d$  must be large enough such that, at very least, less than half of the points in the local neighborhood for any  $t_0$  have significant regional signal  $m$ . The smallest possible value of  $d$  we refer

## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



to as  $d_0$ ; in extraction of background signals,  $d_0$  would be roughly twice the length of the longest regional signal (measured in numbers of measurements). The difficulty we face with this approach is to clearly separate the background signal from the regional signal. As earlier discussed, this is generally an ill-defined problem and can be solved only with additional assumptions on the background signal. Considering this difficulty, we prefer to separate background signals from regional signals by defining the background signal as the estimated smooth curve obtained from the REBS technique using a bandwidth of three month. Such an approach seems reasonable when assuming a regional signal of length shorter than one month and assuming a background signal which varies slowly relative to the regional signal. On the other hand, the bandwidth is short enough to account for possible seasonal effects.

Finally, in order to implement the REBS technique the scale parameter  $\sigma$  (i.e. the measurement noise) needs to be specified. In certain cases,  $\sigma$  can be estimated a priori, e.g., based on the precision of the measurement device. Note that this, however, would neglect the fraction of  $\sigma$  that is due to variability in the background signal. When no a priori information is available,  $\sigma$  must be estimated from the measurements themselves. Since there may be many time points, where the regional signal  $m$  is close to 0, the right side of the residual distribution (the positive residuals) may be long-tailed due to measurements of locally or regionally polluted air masses. Consequently, the scale parameter  $\sigma$  is calculated from the standard deviation of the negative residuals only:

$$\hat{\sigma}_{\text{asd}} = \sqrt{\frac{1}{\#\{j : r_j \leq 0\}} \sum_{i:r_i \leq 0} r_i^2}. \quad (5)$$

In an ordinary (local) least-squares fit or in many REBS applications, the mode of the residuals is at 0. In some applications the mode is below 0 and estimation of the scale parameter using Eq. (5) results in a too large estimate for  $\sigma$ . In these cases all residuals below the mode instead of all negative residuals are used for estimation of  $\sigma$ ,

$$\hat{\sigma}_{\text{masd}} = \sqrt{\frac{1}{\#\{j: r_j \leq \hat{\mu}\}} \sum_{i: r_i \leq \hat{\mu}} (r_i - \hat{\mu})^2}, \quad (6)$$

where  $\hat{\mu}$  is the estimated mode of the residuals distribution. Unfortunately, the estimation of the mode of an empirical distribution is challenging. We use either a nonparametric density estimator as they are described e.g. in Simonoff (1996) and in Fan and Gijbels (1996) or we simply use a histogram with many classes.

In both approaches, the precision of the measuring instrument (e.g., the standard deviation of working standard measurements) can be considered as a lower bound for the estimate of the scale parameter.

To summarize, the REBS technique proceeds as follows:

1. For each observations  $Y(t_i)$ , compute  $\hat{g}(t_i)$  by using the local regression estimator of Eq. (2) with the kernel weights defined by Eq. (3) and robustness weights  $w_r(t_i)=1$ .
2. Use Eq. (5) or Eq. (6) to estimate the scale parameter  $\sigma$  and calculate the robustness weights  $w_r(x_i)$  by applying Eq. (4).
3. For each observation  $Y(t_i)$  compute a new fitted value  $\hat{g}(t_i)$  by using the robust local regression estimator of Eq. (2) with kernel weights defined by Eq. (3).
4. Repeat steps 2 and 3 until convergence, which generally requires about 5–10 iterations. The final fitted values yield the estimated curve  $\hat{g}(t_i)$ .
5. All observations  $Y(t_i)$  with  $Y(t_i) \leq \hat{g}(t_i) + 3\sigma$  are classified as “background” measurements, all other observations are classified as “polluted”.

## 2.2 The uncertainty in the resulting curve

Confidence bands are the common notions to formalize the uncertainty of fitted curves. Classically, they are based on analytical deviations from the asymptotic distribution of

### Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the fitted values. In our case, such an approach is very tedious since we use asymmetric robustness weights. Thus we propose to use the bootstrap approach (Efron and Tibshirani, 1993) which is a general-purpose technique for obtaining information such as confidence bands by simulation. The basic idea is to repeatedly simulate from the residuals new sample sets of residuals and hence compute sets of pseudo-responses. With each set of pseudo-responses a new background signals is extracted. This is repeated  $B$  times. To take into account the temporal spread of the regional signal, we resample from blocks of consecutive residuals. In our setting, the blocks do overlap.

When robust estimates are bootstrapped two problems arise: numerical instability and computational cost. The first problem is due to very poor estimates resulting from the bootstrap pseudo-responses which may contain a higher proportion of regional signal than the original data. The latter problem is due to the complex estimation procedure, which must be used in order to calculate robust estimates.

To overcome these problems, Salibian-Barrera and Zamar (2002) propose the so-called robust bootstrap which is fast and can resist large proportion of outliers in the bootstrap pseudo-responses. Their idea is to bootstrap the pair residuals  $r_j$  and robustness weights  $w_r(x_j)$  simultaneously and use the corresponding robustness weights for the pseudo-responses in a one step iteration of the REBS technique. This idea does, however, not take into account the uncertainty caused by estimating the robustness weights. Hence confidence intervals based on this modified bootstrap idea must be corrected as suggested in Salibian-Barrera and Zamar (2002). As the REBS techniques implies to calculate the corrections at each point  $t_0$ , the computation of these corrections is (too) time consuming. Thus we simplify the correction by taking the experience into account that the uncorrected confidence intervals are about 10 to 20% too small. So we end up enlarging the uncorrected confidence intervals by the factor 1.2. By this modification, we should still get a fair idea of the uncertainty in the curve estimated by the REBS technique. Note that the resulting bootstrap confidence are generally asymmetric due to the asymmetric robustness weights as defined in Eq. (4).

### 3 Experimental

#### 3.1 Measurement site

The high alpine research station Jungfraujoch (3580 m a.s.l.) is located on the main crest of the Bernese Alps, Switzerland. JFJ is part of the Swiss National Air Pollution Monitoring Network (NABEL) and one of the “global” stations of the Global Atmosphere Watch (GAW) programme.

#### 3.2 CO measurements at Jungfraujoch

CO has been continuously monitored since 1996 using commercially available NDIR monitors (APMA-360 and APMA-370, Horiba). Modification of the instrument included drying of the air by a Nafion dryer in split flow mode (Permapure PD-50T-24). The CO instrument is calibrated approximately in monthly intervals using a commercial CO calibration gas referenced against NIST (National Institute of Standards and Technology) SRM (Standard Reference Material) and NMI (Netherlands Measurement Institute) PRM (Primary Reference Material) all being consistent with the WMO-2000 scale. Automatic instrument zero checks were performed every 49 h using zero air. The detection limit for individual one minute values is 20 ppb, the overall measurement uncertainty was estimated to be  $\pm 5\%$  ( $1\sigma$ ) (Zellweger et al., 2000).

In a recent study, Zellweger et al. (2009) compared different CO measurement techniques during a field campaign at JFJ and confirmed the suitability of the NDIR method for CO measurements at this site.

#### 3.3 Baseline determination by the smooth curve fit

The proposed REBS technique, is compared with a method that is similar to the approach of Novelli et al. (2003) and denoted here as the smooth curve fit (see Sect. 1). For the smooth curve fit, the measured data are fitted by the parametric function

## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



$$f(t_i) = a_1 + a_2 t_i + a_3 t_i^2 + \sum_{j=1}^4 [a_{(2j+2)} \sin(2\pi j t_i) + a_{(2j+3)} \cos(2\pi j t_i)] \quad (7)$$

where  $t_i$  is the time of observation  $i$ . The polynomial in Eq. (7) represents the trend, the sum of harmonics are an approximation of the average seasonal cycle. In contrast to Novelli et al. (2003), filtering of the residuals was done in the time domain and not in the frequency domain. Fourier transform requires data with regular sampling periods and cannot deal with missing data. As for many long-term data sets, reasonable replacement of data gaps by interpolation techniques is difficult for the CO time series from Jungfraujoch. Another difference between the smooth curve fit and the method of Novelli et al. (1998) is that the accepted deviation of background measurements from the smooth curve fit is not calculated from all residuals but (similar to the REBS technique, see Sect. 2) only from the residuals that are less than the mode of the residuals distribution.

## 4 Results and discussion

### 4.1 Identification of background measurements

Background concentrations might be understood as “the concentration of a given species in a pristine air mass in which anthropogenic impurities of relatively short lifetime are not present” (Calvert, 1990). Consequently, background measurements should be normally distributed with a mode representing the mean background concentration. Therefore, the left side of the distribution of the residuals from a baseline fitting technique (the residuals below the mode of the distribution) should approximately follow a Gaussian distribution as well. As indicated by Balzani Lööv et al. (2008), the above definition of background conditions is only valid for long-lived compounds, for

## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



compounds with short or medium lifetime a generally applicable definition of background does not exist.

Figure 1 shows the histogram of the residuals for the REBS technique applied to hourly CO measurements at Jungfraujoch for the period from 1996 to 2009. CO has an average lifetime of about 2 month that strongly varies from tens of days to up to one year depending on season and location and is therefore not well-mixed in the troposphere (Jacob, 1999; Holloway et al., 2000). Nevertheless, the left side of the residuals distribution follows approximately a Gaussian distribution, the estimate for the scale parameter  $\sigma$  is 15.6 ppb. As mentioned in Sect. 1, the scale parameter  $\sigma$  is an upper limit for the precision of the instrument. However, the obtained value for  $\sigma$  is considerably larger than the random uncertainty of the NDIR instrument (4.2 ppb), which was determined from the standard deviation of repeated zero air measurements (Zellweger et al., 2009). The larger scale parameter  $\sigma$  might be a consequence of the deviations from the concept of background conditions mentioned above: the relatively short lifetime of CO mainly due to oxidation by OH leads to a latitudinal gradient for CO, and therefore to a dependence of the background concentration at JFJ on the air mass origin.

Classification of background measurements  $Y(t_i)$  by  $Y(t_i) \leq \hat{g}(t_i) + 3\sigma$  (Sect. 2) leads to an overestimation of the number of background measurements and to a small bias in estimated baseline curves. This can be seen from Fig. 1, where the frequencies of the residuals that are larger than the mode of the residuals distribution are higher than expected from the fitted Gaussian distribution. A possible way to adjust for this bias could be to randomly select an appropriate number of observations in reasonably narrow bins of the frequency distribution and to treat them as “polluted” measurements. Such a correction has not been done within this work.

The time series of CO background measurements at JFJ as identified by the REBS technique and the smooth curve fit are shown in Fig. 2. In Table 1, the number of hourly CO measurements at JFJ from 1996 to 2009 that are classified as “background” and as “polluted” by the REBS and the smooth curve fit are listed. The classification by the two

**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



methods is very similar, although the REBS classifies some observations as “polluted” that are considered as representative for background conditions by the smooth curve fit. This tendency can be explained by the standard deviation of the residuals below the mode of the residuals distribution, which is 18.3 ppb for the smooth curve fit and therefore 2 ppb larger than the estimated scale parameter  $\sigma$  for the REBS.

The differences in the classification of background measurements have a rather small impact on the estimation of average background CO concentrations. Annual background CO concentrations for the 1996 to 2009 period obtained with the REBS technique are on average 1.8 ppb smaller (range 0.6 to 3.8 ppb) than those obtained from the smooth curve fit.

It should be noted that the enhancements in annual background CO at Jungfraujoch in 1998 as well as in 2002 and 2003 (Fig. 2) are probably due to the impact of emissions from widespread boreal forest fires reported by Novelli et al. (2003) and Yurganov et al. (2005).

## 4.2 Estimation of baseline curves

Figure 3 shows the estimated baseline curve for the REBS technique including the 95% bootstrap confidence band. As indicated in Sect. 2.2, the bootstrap confidence interval is asymmetric around the estimated baseline, the average width of the confidence band ranges from  $-3.5$  ppb to 3.8 ppb. As the bootstrap confidence band indicates, some of the wiggles in the estimated background signal may not be statistically significant. This may also mean that the true background signal is somewhat smoother than estimated by the REBS. Hence one might be tempted to increase the bandwidth  $h$ . However, a large bandwidth  $h$  has the disadvantage of oversmoothing true temporal variability.

For comparison, the baseline curve obtained from the smooth curve fit is also included in Fig. 3. The baseline curve derived from the REBS technique is generally somewhat lower than the baseline curve from the smooth curve fit. There is good agreement between the two approaches during the warmer season when background CO concentrations are lowest due to oxidation by OH, and considerable disagreement

during the cold period when background CO concentrations are highest. These differences would be of importance when emission estimates are performed using techniques as described e.g. by Reimann et al. (2005), Simmonds et al. (2001) and Grealley et al. (2007) that are based on concentration above background estimates. However, the observed differences can be explained by the different methodical concepts applied: the REBS is a purely non-parametric technique that is capable to follow any long-term trend and seasonal variation. On the other hand, the smooth curve fit is less flexible: the parametric model fit given by Eq. (7) gives estimates for the long-term trend and the average seasonal variation of the observations. Superposition of the smoothed residuals allows for adjustment for temporary deviations from the long term trend and for deviations from the average seasonal cycle, however, the main features of the course of the underlying parametric fit remains.

The linear trend of the CO background concentration was determined from regression of the annual means of the identified CO background concentrations against time. The average annual change for the 1996 to 2009 period as determined from the REBS filtered data is  $-2.1 \pm 1.3$  ppb/yr, data filtering using the smooth curve fit results in  $-2.3 \pm 1.3$  ppb/yr, a slightly larger although not statistically significant different decrease of background CO (see also the study by Zellweger et al., 2009 for a discussion of the trend of CO at JFJ for the 1996 to 2007 period). Note that the linear negative trend of unfiltered hourly CO measurements for this time period is considerably larger ( $-2.9 \pm 1.5$  ppb/yr) due to more severe and more frequent pollution events during the first years of the considered time period (see Fig. 2).

## 5 Applicability of the REBS

As discussed in Sect. 4.1, trace gases (e.g. CO) can show latitudinal gradients due to the dependence of emissions and sinks on latitude. If strong latitudinal transport events reach a sampling location, observations might therefore not be representative of the latitudinal background concentration at the considered sampling site. Statistical

### Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Extraction of background concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



filtering methods including the REBS and most meteorological filters cannot correctly cope with the effect of latitudinal gradients. For the REBS, it is suggested that the residuals distribution as shown for CO at JFJ in Fig. 1 is used for judgment of the applicability of the REBS for the time series of interest. The left hand site of the residuals distribution should approximately follow a Gaussian distribution. Obvious deviations from this requirement indicate a significant impact of latitudinal transport or other processes leading to observations that are well below the background concentration at the sampling site. Observations well below the background concentration receive too high weights (see Eq. 4) and consequently lead to a baseline estimation that is biased downwards.

The REBS approach was recently extended by including the latitude of air mass origin taken from back-trajectories as a second dimension in the local regression. This extended data filtering concept for estimation of background concentrations and latitudinal gradients of trace gases was denoted 2D-REBS and will be subject of a forthcoming publication.

A final issue to consider is missing data. The REBS technique can handle data gaps, whereas estimation of missing values and data interpolation is not recommended. In contrast to other techniques like running mean estimations, locally weighted regression as used in the REBS has no serious bias problems near the boundaries of data (Hastie et al., 2001).

**6 Conclusions**

A statistical method based on robust local regression is introduced. The presented REBS technique was applied for identification of background CO measurements at the high-alpine background site at Jungfraujoch, the results were compared to those from a more common approach denoted here as the smooth curve fit.

Both methods resulted in similar selection of background measurements and good agreement in linear trend estimates, but in considerable differences in the baseline

**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



curves. The latter disagreement is explained by conceptual differences of the two methods. As a purely non-parametric approach, the REBS technique is more flexible than the smooth curve fit and can reproduce any long-term trend and highly variable seasonal cycles. In contrast, the smooth curve fit has limited capabilities to follow variations in the seasonal cycle. Moreover, the smooth curve fit can only be successfully applied to species with a long-term trend that can be described by a polynomial approach as expressed by Eq. (7). If this is not the case, it might be difficult to find and to include an appropriate parametric function for estimation of the long-term trend. The REBS technique is certainly computationally more expensive and conceptually more complicated than the smooth curve fit. The above mentioned advantages of the REBS should, however, outweigh these disadvantages in many applications.

*Acknowledgements.* The authors gratefully acknowledge the support from the International Foundation High-Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG), and the custodians at the JFJ research station for their hospitality. The CO measurements at JFJ are part of the Swiss National Air Pollution Monitoring Network (NABEL) and supported by the Swiss Federal Office for the Environment (FOEN).

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## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## Extraction of background concentrations

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

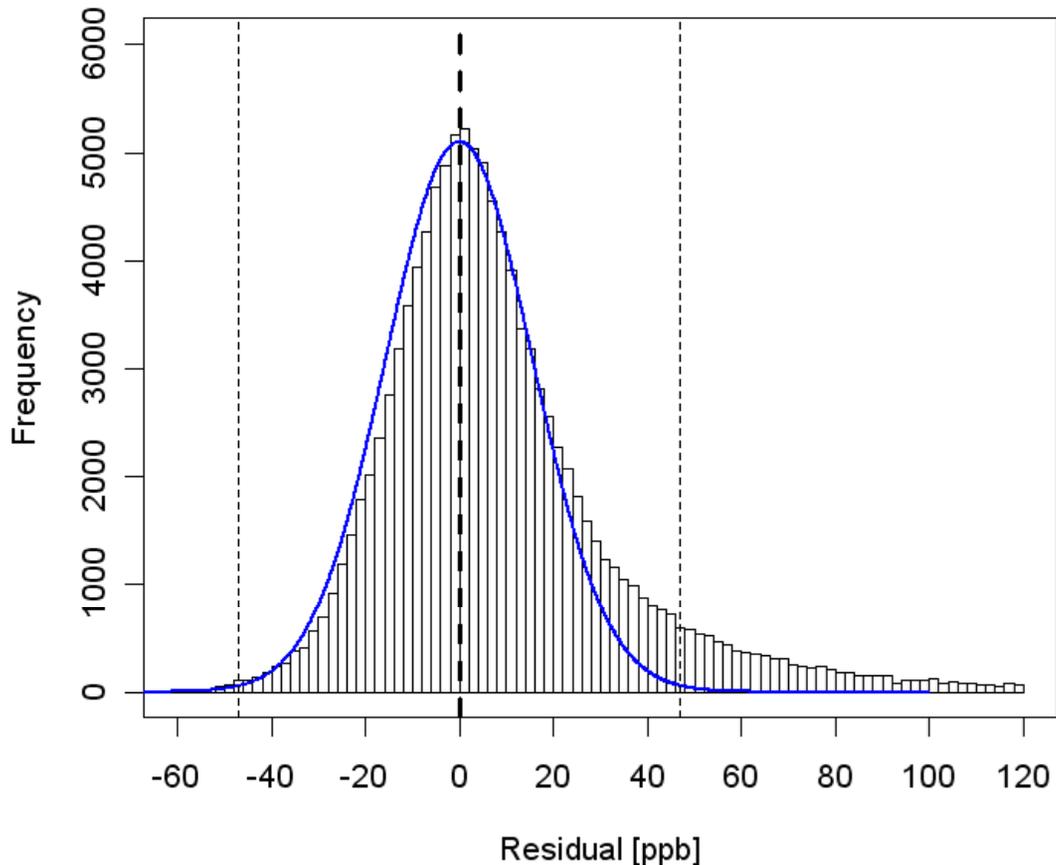
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Interactive Discussion



**Table 1.** Contingency table of the classification of the hourly CO values measured at Jungfraujoch from 1996 to 2009 ( $n=111\,668$ ) derived from the REBS technique and the smooth curve fit.

		smooth curve fit	
		background	polluted
REBS	background	102 405 (91.7%)	0 (0.0%)
	polluted	3188 (2.9%)	6075 (5.4%)



**Fig. 1.** Histogram of the residuals derived from application of the REBS technique to hourly CO measurements at Jungfraujoch. The estimated scale parameter  $\sigma$  is 15.6 ppb. The thick vertical dashed line indicates the estimated mode of the residuals distribution, the two thin vertical dashed lines denote the  $\pm 3\sigma$  range. The blue line is a Gaussian distribution fitted to the left side (residuals below the mode) of the residuals distribution.

**Extraction of background concentrations**

A. F. Ruckstuhl et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

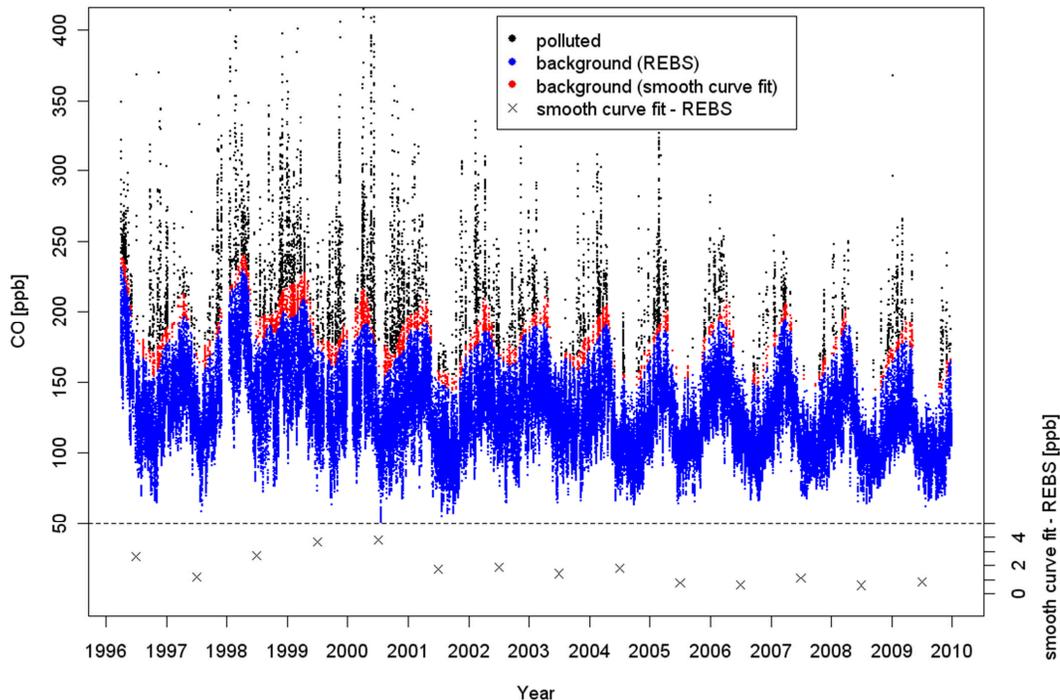
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Interactive Discussion



**Extraction of  
background  
concentrations**

A. F. Ruckstuhl et al.



**Fig. 2.** Measured CO during impact of regionally polluted air masses (black points) and background CO concentrations at Jungfraujoch for the 1996–2009 period as identified by the REBS technique and the smooth curve fit. The black crosses indicate the difference of annual background concentrations obtained by the two data filtering methods.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

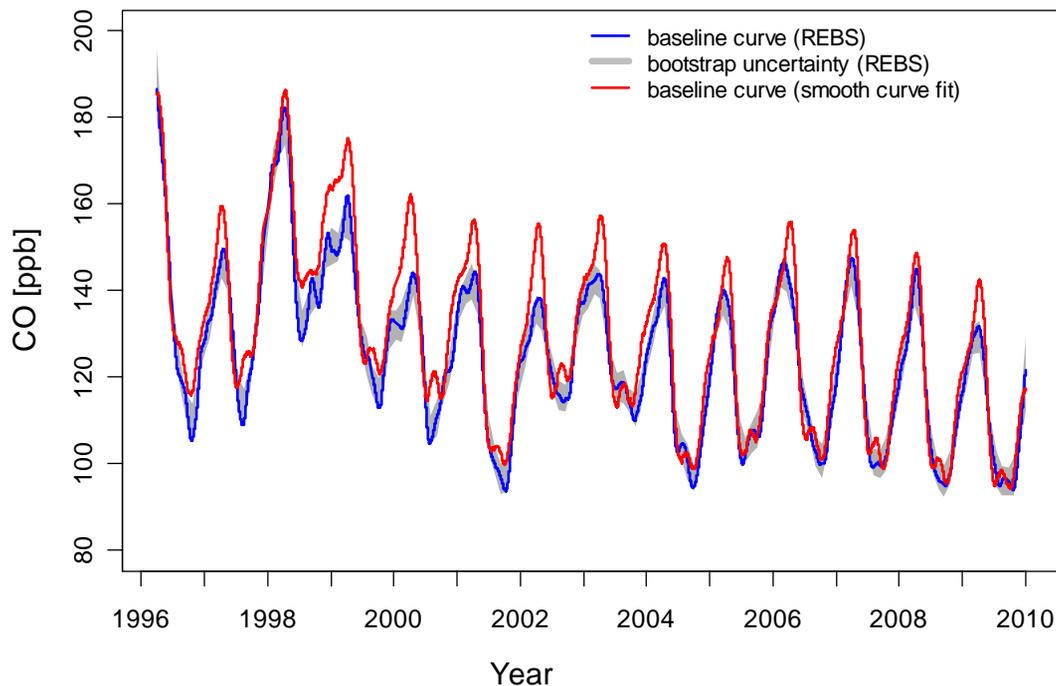
Printer-friendly Version

Interactive Discussion



**Extraction of background concentrations**

A. F. Ruckstuhl et al.



**Fig. 3.** Baseline curves for CO at JFJ (1996 to 2009) obtained with the REBS technique (including the bootstrapped 95% confidence band) and the smooth curve fit.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)