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METHODOLOGICAL DEVELOPMENT OF THE CONDITIONAL SAMPLING METHOD. PART II: QUALITY CONTROL CRITERIA OF RELAXED EDDY ACCUMULATION FLUX MEASUREMENTS

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Abstract. Determination of biosphere–atmosphere exchanges requires accurate quantification of the turbulent fluxes of energy and of a wide variety of trace gases. Relaxed Eddy Accumulation (REA) is a method that has received increasing attention in recent years, because it does not require any rapid sensor for the scalar measurements as the Eddy Correlation method (EC) does. As in all micrometeorological studies, REA measurements in the atmospheric surface layer are valid under some restrictive conditions so as to be representative of a specific ecosystem. These conditions are the homogeneity of the underlying surface, stationary and horizontally homogeneous turbulence. For most experiments these conditions are not fully satisfied. Data uncertainties can also be related to not fulfilling the method principles or to the technical characteristics of the REA system itself. In order to assess REA measurement quality, a methodological approach of data analysis is developed in this study. This methodological analysis is based on the establishment of criteria for data quality control. A set of them, deduced from the mean and turbulent flow, are called ‘Dynamic criteria’ and are designated to control the stationarity and homogeneity of the w function and the validation of Taylor’s hypothesis. A second set (‘REA operational criteria’) is designed to check the sampling process and, more precisely, the homogeneity of the negative and positive selection process throughout the sampling period. A third set of criteria (‘Chemical scalar criteria’) concerns the scalar measurements. Results of the criteria application to data measured at two different experimental sites are also presented. Cut-off limits of criteria are defined based on their statistical distribution and shown to be specific for each site. Strictness of each criterion, defined by the percentage of flagged samples, is analysed in conjunction with the meteorological conditions and atmospheric stability. It is found that flagged samples mainly correspond to neutral and stable nocturnal conditions. During daytime, nearly free convection conditions can also yield poor quality data.

Keywords: Atmospheric stability, Data quality control criteria, Relaxed eddy accumulation method, Turbulent functions.

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1. Introduction

The relaxed eddy accumulation (REA) method allows flux measurements for many scalars for which analyser response time is not rapid enough to apply the eddy correlation method (EC) (Businger and Oncley, 1990). The conditional sampling technique and equations on which the REA is based are summarised in a companion paper (Fotiadi et al., 2005). Simulations of REA operation, also presented in Part I, quantify sensitivities in flux estimate related to (1) the statistical characteristics of turbulent functions through the skewness and the kurtosis; and (2) the selection algorithm with application of a threshold, time lag and filtering on the vertical velocity (w) signal. All of these tests were made using generated functions. The goal was to emphasise the method's sensitivity to a number of parameters that can be controlled, or checked afterwards.

All of the micrometeorological methods used to estimate turbulent exchanges over large homogeneous areas are based on the same hypothesis of horizontal homogeneity of the surface and stationarity of turbulence. These conditions are well fulfilled in the case of a flat surface with a sufficient fetch, constant roughness length, and with sinks and sources homogeneously distributed on the surface. Under some experimental conditions these assumptions are not fully satisfied, especially when continuous measurements over a long period of time are realised, including diurnal transition and stable conditions. Therefore, once the measurements are made, data control is necessary to guarantee their quality for further use (Foken and Wichura, 1996; Affre et al., 2000). Such a control is relatively simple in the EC method since all the turbulence functions involved are available and can be verified at all post-treatment steps. In that method, all of the information needed to evaluate the calculation precision is available (Mann and Lenschow, 1994). This is not the case for conditional methods. In the REA method, the vertical velocity function, which drives the air sampling process, is the only indicator of the turbulence conditions encountered. Sometimes, low frequency information of the studied scalar is available. Then, data post-treatment bears on the knowledge of beta dependency on air selection (threshold...) and on statistical moments of w . There is no possibility to perform this treatment based on the difference in scalar concentrations between the two reservoirs. Since evaluation of the calculation precision is not accessible, a set of criteria is proposed in order to mark samples collected in questionable conditions.

A methodological analysis for quality assessment of REA measurements has been developed, and is presented here. This quality control is realised through a number of selection-criteria tools. Some of them, applied to the vertical velocity turbulent function, were previously defined for the control of the EC measurements by Affre et al. (2000) and are properly adjusted

to the REA concept. Other selection criteria, specific to the REA method, are established and concern the sampling process itself, through the control of the selection time. At last, even though the turbulent fluctuations of the chemical compound are not available, some criteria are defined from the low frequency concentration acquired by an analyser. The chemical concentration stationarity and also the REA system and the analyser functioning are controlled.

Sample selection is performed based on cut-off limits of these various criteria. Evidently, the cut-off limits are site dependent and, as a consequence, a theoretical threshold can rarely be established (except for the turbulence intensity, the detection limit, and the concentration centrality). From there, the cut-off limit for each criterion has been established based on the distribution of this criterion throughout the campaign. Samples were flagged when criteria were out of the range (more than 3 standard deviations, or skewness in absolute value higher than 1).

As an illustration, the application of this methodology to control NO_x fluxes estimated by the REA method is analysed. Since this study is a methodological approach destined to be applied to experimental data, first the experimental sites where the data were collected are presented. The two sites are characterised by two different homogeneous ecosystems. Then, the data selection criteria are defined and applied to the experimental data. The strictness of each criterion, defined by the percentage of flagged samples, is analysed in conjunction with the meteorological conditions and atmospheric stability.

2. Field Sites, Experimental Set-up and Acquirement Description

Data were collected using the same REA system during two experiments (“La Crau” and “Maize”) carried out in the framework of the experimental campaign ESCOMPTE 2001, June–July 2001, in the Marseille region in south-eastern France (Cros et al., 2003).

The vertical wind velocity component was measured with a tri-axial ultrasonic anemometer (Young, model 81000, 0.15 m path length). All w time series were sampled at 5 Hz over a sampling period of 15 min. Teflon solenoid valves (ASTI, model EV 360) with a response time of approximately 10 ms were used for the sample selection. Tubing between the inlet and the first valve was of 0.22 m length and 4 mm inner diameter. Given the pump rate of 1.7 l min^{-1} , the transit time in the tubing before the first selection valve is about 0.1 s. The sonic signal (w) was acquired by an 8-bit analog-to-digital conversion board, and after a lowpass filtering at 5 Hz cut-off frequency was applied. A threshold on the w signal, equal to $0.1\sigma_w$ for the “La Crau” experiment and $0.2\sigma_w$ for the “Maize” was used (where

σ_w is the standard deviation of the w function of the previous 15 min sample). For the “Maize” site, NO_x fluxes result from the competition between the two opposite processes of NO emissions from soil and NO_2 deposition onto the vegetation and ground. This competition can result in smaller deposition fluxes, or even in reverse (positive or emission) fluxes when compared to the “La Crau” site where only deposition takes place. Increasing the w threshold allows a better sensitivity on the $\Delta X = \overline{X^+} - \overline{X^-}$ determination (with $\overline{X^+}$ = mean concentration of updrafts and $\overline{X^-}$ = mean concentration of downdrafts). The threshold was centred around the mean vertical velocity, \overline{w} , of the previous 15-min sequence.

The REA system was complemented by turbulent energy fluxes (momentum, sensible and latent heat fluxes) measured by the EC method at 6 m and 8 m height for “Maize” and “La Crau” sites, respectively. The sampling frequency of the ultrasonic anemometer (Gill instruments, model 1012R2) and the Licor (6262) sensor used for the energy fluxes was 21 Hz, with a 15-min averaging period.

The two experimental sites are described in the following sections through the surface characteristics (soil and vegetation), and the dynamic conditions with the associated NO_x concentration levels. Roughness length, also presented, is deduced from the drag coefficient calculated through the ratio of the friction to mean wind velocity (u_* / u) for neutral conditions.

2.1. “LA CRAU” SITE

The experimental site is located in the semi-arid part of La Crau plain (43°34' N, 04°49' E, 10 m elevation). La Crau plain, lying to the west of Marseille, occupies a surface area of 50,000 ha whose major part is cultivated. The semi-arid part of the plain covers a surface of 11,500 ha and consists of a flat and horizontal (maximum slope is lower than 0.3% in direction 040°), uniform, and with almost bare soil composed of clay with pebbles. Vegetation, pasture type, is very sparse. The estimated roughness length is about 15 mm. The site is characterised by a uniform fetch in all directions (at least 1 km). The experiment was conducted between May 15th and June 7th, 2001. A set of 1450 samples of 15-min duration collected at 6 m height was analysed in this study. Figure 1a presents in the same plot, the wind rose and the mean wind speed for each direction. Figure 2a shows the mean concentration of NO_x as a function of the wind direction. Two prevailing wind directions can be observed: (1) a regional wind of north-north-west direction (340°) called the ‘Mistral’, with relatively high speeds (7 m s⁻¹, accompanied by low NO_x concentrations of 2 ppb (Figure 2a); and (2) a south-east wind (160°), reinforced by the sea-breeze circulation (4–5 m s⁻¹). This regime is generally related to pollution transport from the industrial area situated about 20 km upwind (5 ppb

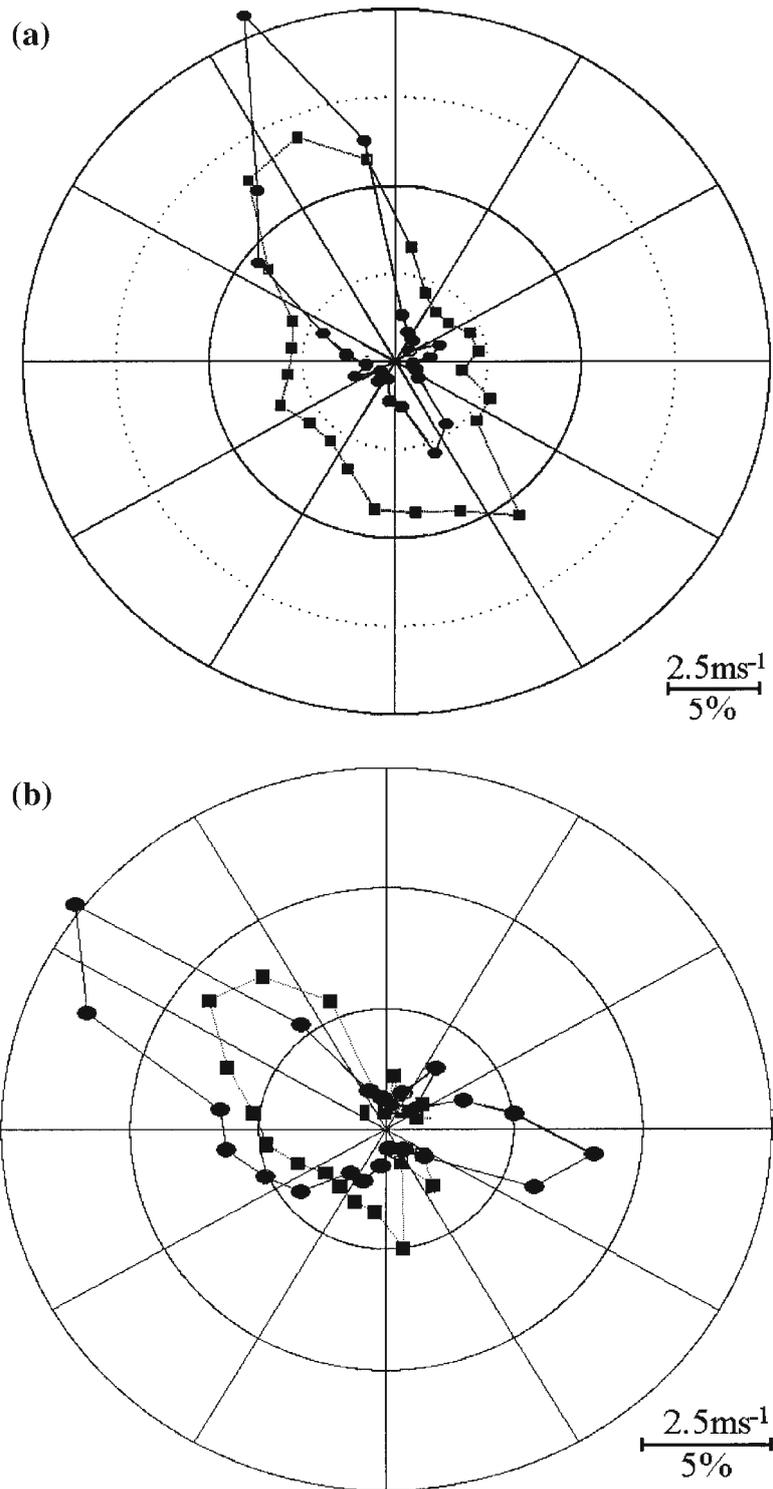


Figure 1. Wind roses for “La Crau” (a) and “Maize field” (b) sites. Dark circles represent the frequency of wind direction, with coordinates of 10 and 20% for (a) and 5, 10, and 15% for (b). Solid squares represent the mean wind velocity related to each direction, with coordinates of 5 and 10 m s⁻¹ for (a), and 2.5, 5, and 7.5 m s⁻¹ for (b).

as indicated by the mean values reported on the Figure 2a but individual samples can reach 20 ppb).

2.2. “MAIZE” SITE

The REA system was set up in a 13-ha maize field at Meyrargues (43°39' N, 5°32' E, 195 m elevation), in the Durance valley. The fetch exceeded 200 m in all directions apart from the south, where the field boundary was approximately 100 m distant. The surrounding area was cropland. The terrain in the area is flat and horizontal (maximum slope lower than 0.33% in direction 110°). Soil at the site was predominantly fine clay and silt (clay = 30.5%, silt = 53.6% and sand = 15.9%). The maize canopy height rose from 0.5 m at the beginning, up to 1.2 m at the end of the experiment. Leaf area index (LAI) varied from 1 to 2.6. The averaged roughness length estimated for the whole experiment period is about 0.3 m. The measurements were carried out from June 12 to June 30, 2001 during a rain-free period. The ultrasonic anemometer was mounted 4.5 m above the ground surface and a set of 1669 samples of 15-min length was collected and analysed for the present study. The “Maize” site is subject to the same wind regimes as the “La Crau” one, with deviations of about 35° in the cyclonic sense due to the orography and the orientation of the valley (Figure 1b). The ‘Mistral’ has then a north-west direction (310°) and a speed of about 4 m s⁻¹. It is associated with the lower NO_x concentration levels (1.5 ppb in average) observed at this site (Figure 2b). The second main regime is related to an east wind (100°) with low speeds (1.5 m s⁻¹), related to mean NO_x concentrations of 3 ppb. Contrary to the “La Crau” site, and except for the Mistral direction, the NO_x concentrations at the “Maize” site are relatively homogeneous according to the wind direction, and do not present sectors of pronounced pollution. It should be noticed that, although these concentrations are mean values over 15 min, they exhibit a high variability. Indeed, the ratios of the standard deviation to the mean concentration can reach 1 for both sites.

3. Criteria Presentation

This analysis follows a triple approach. First, it concerns mean flow and turbulence characterisation (‘Dynamic’ criteria), focused on the vertical wind velocity component, as w is the guiding factor of the conditional sampling, and the only turbulent function involved in REA measurements. Then, a second set of ‘REA operation’ criteria, associated with the sampling process itself, is established. Finally, other ‘Chemical Scalar’ criteria, regarding the scalar measured, are constructed using the concentration

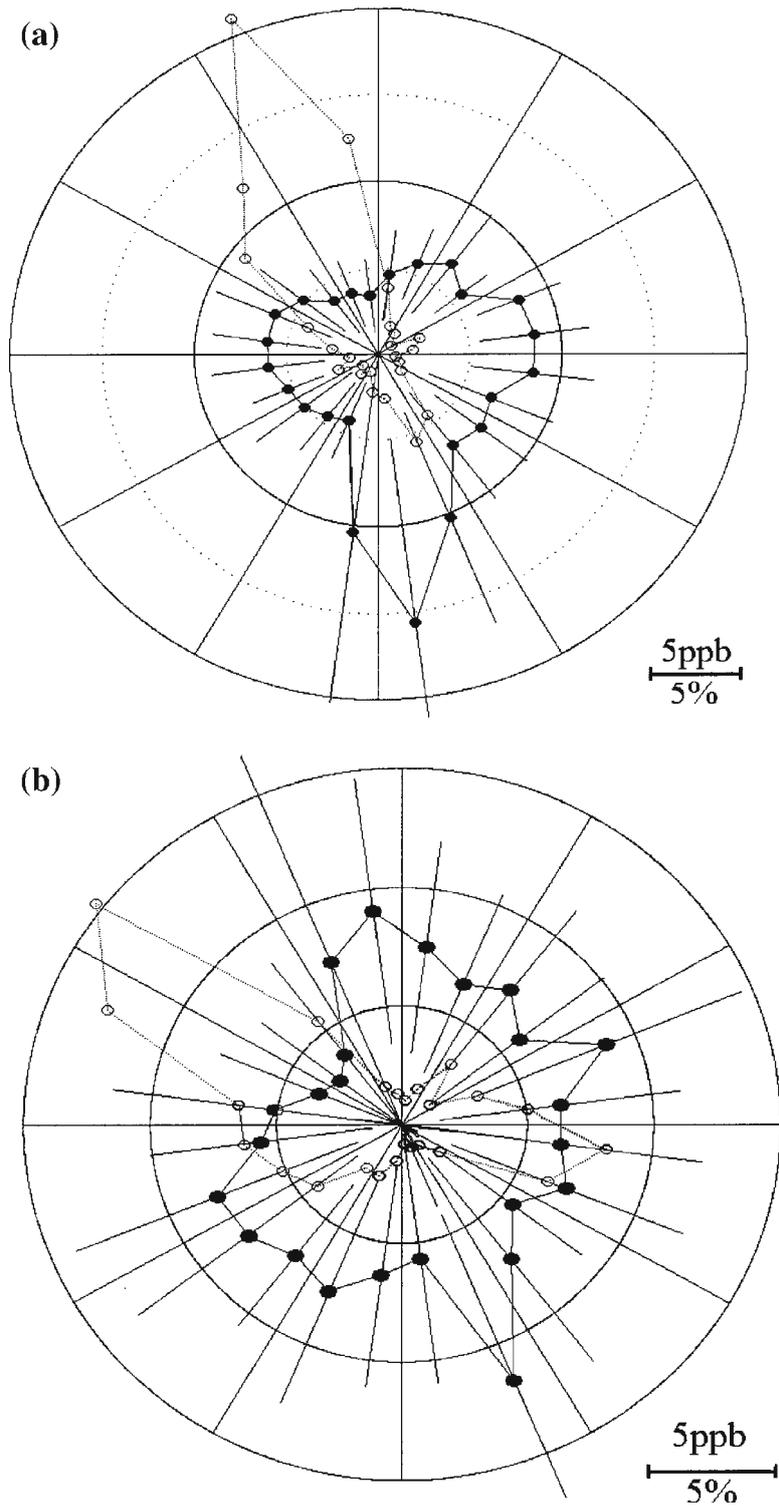


Figure 2. NO_x concentration as a function of wind direction for “La Crau” (a) and “Maize” (b) sites. Circles represent the frequency of the wind direction (scale is the same as in Figure 1). Solid circles represent the mean ambient NO_x concentration. Circle coordinate are 10 and 20 ppb for (a), 5, 10, and 15 ppb for (b). Radial bars centred on solid circles represent the concentration standard deviation.

function. This low frequency function is recorded during the REA sampling period.

The mean diurnal variation of each criterion is presented for the “Maize” experiment. These graphs are established after eliminating the extreme values based on cut-off limits presented and discussed in Section 4.

3.1. ‘DYNAMIC’ CRITERIA

A study of the turbulent processes is made from the statistical characteristics of the w distribution and the analysis of some spectral characteristics.

3.1.1. *Third- and Fourth-Order Moments of w*

Most of the time, distributions of measured turbulent functions deviate from the Gaussian with third- and fourth-order normalised moments (skewness and kurtosis) different from 0 and 3, respectively. It was shown (Fotiadi et al., 2005) that a skewness different from zero implies variations in the REA coefficient (β) with subsequent influence on the flux estimate. On the other hand, the kurtosis has a weak impact on β values, but has a considerable effect on the distribution of points discarded by the threshold application (Fotiadi et al., 2005). Additionally, high absolute values of these two parameters could indicate the presence of noise (kurtosis) and extreme peak values (skewness) in the studied turbulent variable. Both skewness and kurtosis are used as criteria for data quality. Their mean diurnal variation illustrates well the turbulence diurnal cycle (Figures 3a and b). In fact, skewness and kurtosis have opposite behaviour. For the whole measurement period, distributions of w exhibit a positive skewness, which increases up to 0.4 at midday, and tends to zero in the middle of the night. The kurtosis is close to 3 a little earlier in the morning, and, inversely during the night, the turbulent fluctuations are flattened out (kurtosis varies from 4 to 4.5).

3.1.2. *Dissipation Length and Time Scales*

There are several ways of estimating characteristic scales of the largest turbulent eddies. Two of them are currently used: the wavelength corresponding to the maximum of the energy spectrum (λ_m) known as the spectral length scale, and the dissipation length scale (L_ε) (Equation (1)), which represents the size of the largest eddies at the beginning of the spectrum inertial sub-range. This latter parameter is used as a data selection criterion to investigate any possible disturbance in the measurement process. The value of L_ε tends to be very small in the presence of noise in the w function. Conversely, large values are symptomatic of laminar or gravity wave flows. Note that the dissipation scale can be alternatively expressed in time units $T_\varepsilon(s)$, conversion being made according to the Taylor’s hypothesis, and using the mean horizontal wind speed. At the measurement height,

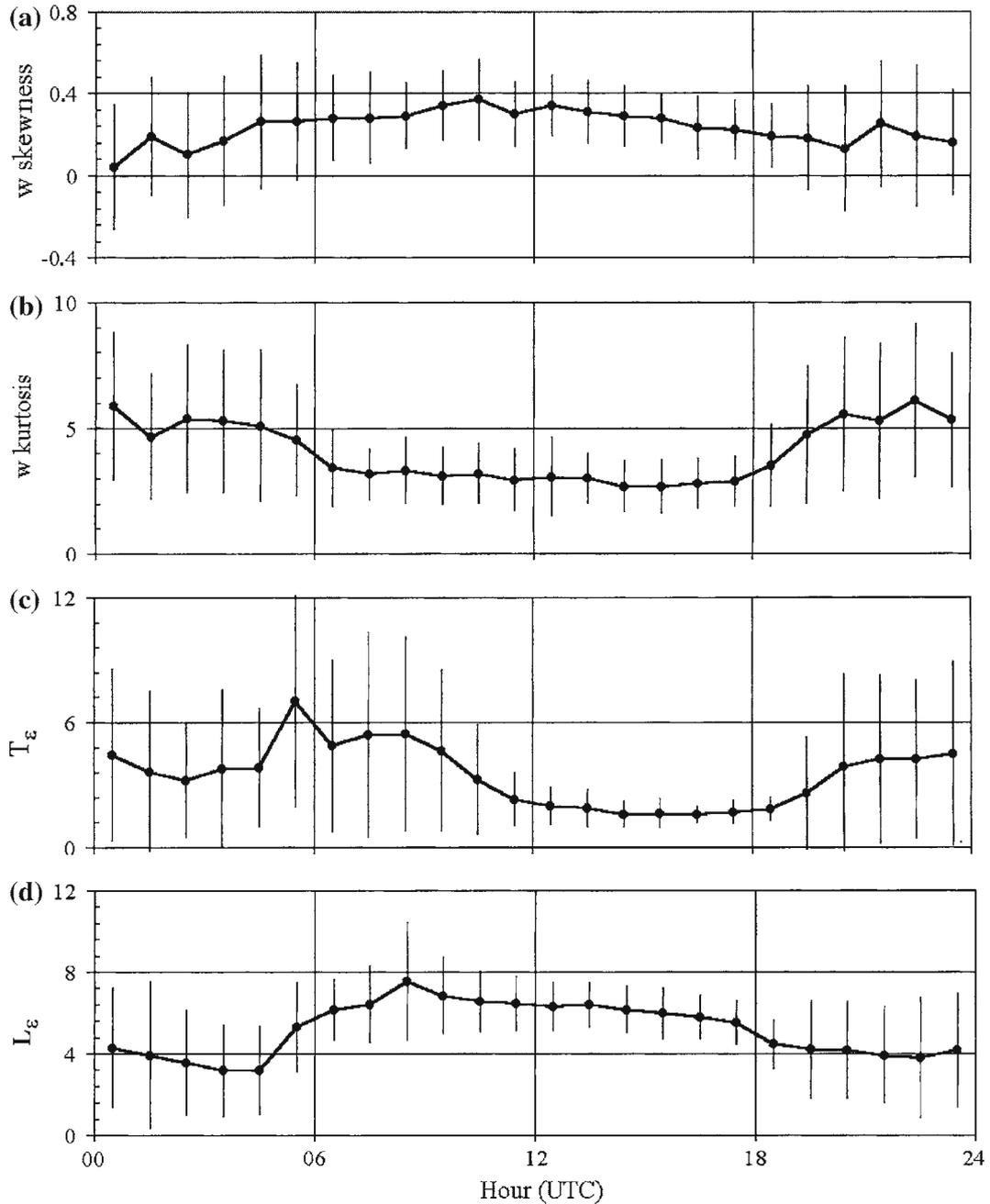


Figure 3. Mean diurnal variation of the ‘Dynamic’ and ‘REA operation’ criteria calculated for the “Maize” site experiment: (a) skewness; (b) kurtosis; (c) dissipation time scale T_ϵ (in seconds); (d) dissipation length scale L_ϵ (in meters); (e) CRS_{-w} , (f) turbulence intensity; (g) mean time of positive selection; and (h) CRS_{-t+} . Vertical bars represent the standard deviation around the mean value indicated by the solid circle.

the dissipation length scale is of the order of a few metres, and the dissipation time scale is on the order of a second. Using the dissipation time scale (T_ϵ) allows the mean horizontal wind velocity to be taken into account and, thereby, to detect free convection conditions.

The dissipation length scale is expressed (Hunt et al., 1985) as a function of the variance σ_w^2 and of the structure function coefficient, C_w^2 , of the vertical velocity:

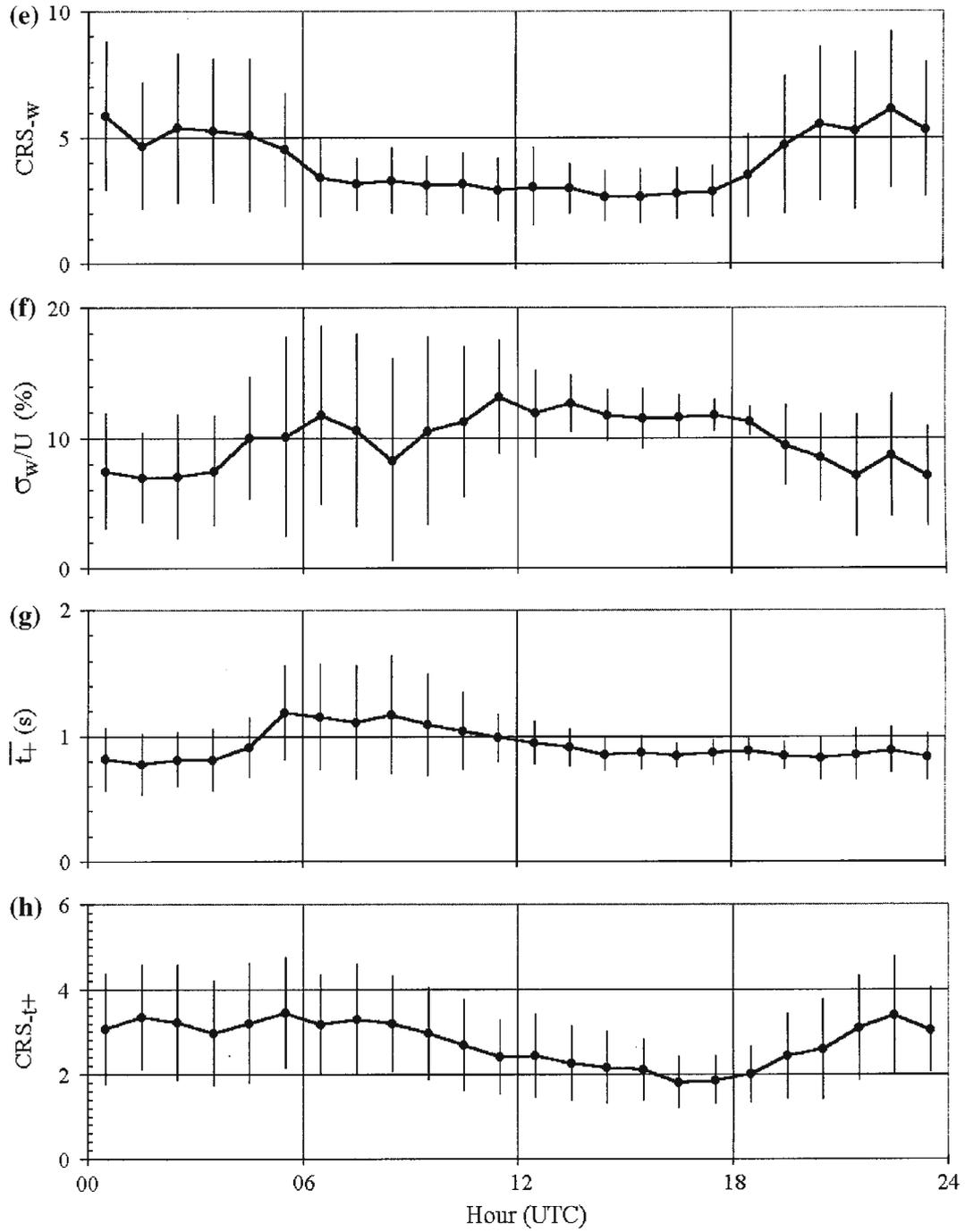


Figure 3. Continued.

$$L_\varepsilon = \left[\frac{2\sigma_w^2}{C_w^2} \right]^{3/2}. \quad (1)$$

C_w^2 is deduced from the linear part of the structure function $D_w(\tau)$ where τ is the time lag between two measurements. Actually, for short lags, the structure function appears as a straight line with a $2/3$ slope, corresponding to the inertial sub-range portion of the spectrum described by the $-5/3$ power law. Then:

$$D_w(r) = C_w^2 r^{2/3}. \quad (2)$$

Figures 3c and 3d show the averaged diurnal variation of the dissipation length and time scales. Even though L_ε is high during the day (6 m at 1500 UTC), at the same time, T_ε is low (2.4 s at 1500 UTC) emphasising in that way the role of the mean wind.

3.1.3. Criterion Relative to w Stationarity (CRS_w)

Turbulence stationarity is one of the fundamental hypotheses that should be fulfilled when determining turbulent fluxes. The presence of low frequencies, which usually are not of local turbulent origin but can be induced by the constraint of large-cell circulations or mesoscale events in the atmospheric boundary layer (ABL), implies longer scale processes in the turbulent fluctuations. They can yield significant disturbances in flux evaluation. For this reason, a criterion related to stationarity (CRS_w) is defined, which also defines the turbulence homogeneity. For that, the contribution to the w variance is studied along the sample. This approach is similar to the one defined by Affre et al. (2000) for fluxes. Given that the sample length is large when compared to the spectral length (λ_m) the homogeneity of the contribution to the w variance along the sample is defined by the integral function as follows:

$$f(t) = \int_0^t w'(\tau)^2 d\tau. \quad (3)$$

This integral function increases monotonically, in contrast to the covariance which can have a changing sign for the slope. The variance can be defined as:

$$\sigma_w^2 = \frac{1}{T} [f(t_2) - f(t_1)], \quad (4)$$

where T is the sample duration. This estimation has a low sensitivity to the limits $f(t_1)$ and $f(t_2)$ when the function $f(t)$ slightly differs from a straight line.

The criterion presented here defines the sample homogeneity. This is done by calculating the mean slope of the least mean square regression line, which corresponds to a statistical evaluation of the variance σ_{wstat}^2 . It takes into account all the function evolution, and not only the extreme points as for the “classical” variance calculation. The homogeneity is quantified by evaluating the standard deviation σ_{wreg} between $f(t)$ and the regression line. The CRS_w , expressed as a percentage, is then defined as the ratio of the standard deviation σ_{wreg} to the w variance,

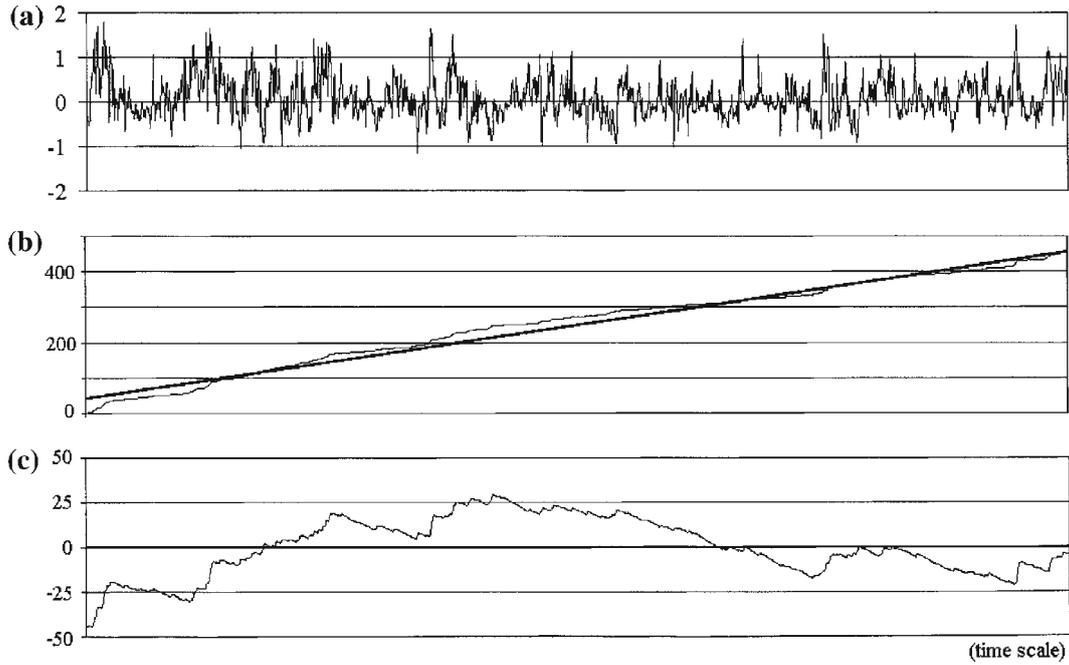


Figure 4. Illustration of the CRS criterion. (a) timeseries of $X(t)$; (b) integral function $f(t)$ of the $X(t)$ square, and linear drift fitted on; and (c) $f(t)$ detrended function.

$$CRS_{-w} = 100 \frac{2\sigma_{wreg}}{[f(t_2) - f(t_1)]} \approx 100 \frac{2\sigma_{wreg}}{T\sigma_{wstat}^2}. \quad (5)$$

It represents the error in the variance evaluation, and the higher is the CRS_{-w} , the less homogeneous is the sample (Figure 4). CRS_{-w} tends to zero (small difference between the integral function $f(t)$ and the linear regression) for a linear $f(t)$ function, and increases with $f(t)$ non-linearity. For oscillating $f(t)$ functions, CRS_{-w} is high, and the ratio L_ε/λ_m is likely to be small. For the ‘‘Maize’’ site (Figure 3e), the most stationary functions were collected during daytime, with a CRS_{-w} about 3%. Night samples are less homogeneous, since the CRS_{-w} reaches 6% on average, and presents a large standard deviation, reaching 2.5%.

3.1.4. Turbulence Intensity

Turbulent fluxes are referred to a surface area integration but they are actually measured at a fixed point in space over a time integration period. The approximation of turbulence measurements as a function of time (t) by their corresponding measurements in space (x) is made using the mean wind velocity and is based on the Taylor’s hypothesis or ‘frozen’ turbulence approximation. According to Lumley and Panofsky (1964) and Wyngaard and Clifford (1977), turbulence intensity, defined as the ratio of the turbulence (σ_u or σ_w) to the mean wind speed (u), can be used as a criterion to validate Taylor’s hypothesis. This hypothesis is applicable to flow with low turbulence intensity.

If the Taylor's hypothesis is not fulfilled, the possible acceleration and deceleration of the mean flow result in a sampling time not being proportional to the size of the air parcel. In other words, statistics referring to time are different to those referring to space. Moreover, this alteration, in combination with a negative $w'u'$ correlation in the surface layer, implies an underestimation of downward motions and overestimation of upward motions. Affre (1997) showed that, for a turbulence intensity of 50% and a $w'u'$ correlation of -0.6 , the alteration of the time sampling statistics may result in an error in the flux estimation by the EC method of about 20%. For the REA method, error induced in the ΔX generates a disturbance in the flux that must be smaller than the corresponding one induced in the EC flux. The higher the turbulence intensity is, the more the statistic is disturbed by the asymmetry induced on the negative and positive sampling time.

As shown in Figure 3f, the turbulence intensity (expressed as a percentage) is higher during the day (12% at midday) than during the night. The scatter of this value is maximum in the morning between 0600 and 1100 UTC when the boundary-layer height increases, and progressively decreases until 1900 UTC.

3.2. 'REA OPERATION' CRITERIA

3.2.1. *Mean Duration of Negative and Positive Selection*

Data analysis presented so far concerns the w quantity. In order to extend the analysis to the REA method, functions of the time of positive and negative selection, and of the non-sampling period, are reproduced for each w sample by simulating the REA operation. These functions are of 'step' form, being +1 for upward motions (positive sampling), -1 for downward motion (negative sampling) and 0 for the non-sampling period, respectively. From these 'step' functions two time parameters can be defined for each sample: the total time of the positive and negative sampling along a w sample, and the mean duration of positive (t_+) and negative (t_-) selections. The first parameters are strongly dependent on the skewness of the w function (they are not presented here). The second set is calculated as the mean length of positive and negative 'step' functions, respectively, and is more appropriate to evaluating the REA performance. Indeed, these two quantities are of great interest from the technical point of view, since they indicate the mean time of valves opening during the sampling period. For this reason, they are employed as quality criteria for REA. The diurnal variation of the positive time selection is presented in Figure 3g (the representation is quite similar for the negative selection, which is not shown here). It exhibits a maximum between 0500 and 0800 UTC and varies from 0.8 s during the night to 1.2 s at the sunrise. One can note the very low

values of the selection time, which is often lower than 1 s. This remark reveals the importance of the study of the time lag impact on the REA method, discussed in Fotiadi et al. (2005).

3.2.2. Criterion Relative to Stationarity and Homogeneity of Sampling Time (CRS_{-t+})

The internal structure of the time functions of positive and negative selection is now studied to check whether the negative and positive samplings are homogeneous and equidistributed along the considered sampling period. In fact, non-equidistributed selection combining with linear or a more complicated drift of the chemical function would lead to a distorted difference between the two reservoir concentrations. In order to control the homogeneity and stationarity, a criterion (CRS_{-t+}) analogous to CRS_{-w} , is defined:

$$CRS_{-t+} = 100 \frac{2\sigma_{t_{reg}}}{T\sigma_{t+}^2}, \quad (6)$$

where $\sigma_{t_{reg}}$ is the standard deviation of the differences between the t_+^2 integral function and the linear regression line, and σ_{t+} is the mean standard deviation of the step function t_+ previously defined. Tests made for the timeseries of negative selection time show that CRS_{-t-} is similar to the one of the positive selection time. Consequently, only the CRS_{-t+} is used as a criterion of the homogeneity of selection time. Even though the CRS_{-w} (Figure 3e) shows that the most homogeneous samples correspond to a long time period between 0800 and 1800 UTC, the diurnal evolution of the CRS_{-t+} (Figure 3h) has a minimum value (1.8%) at 1700 UTC, corresponding to the best selection conditions.

3.3. 'CHEMICAL SCALAR' CRITERIA

As the REA method is often applied to measure turbulent fluxes of chemical compounds, it is then necessary to establish criteria to control the scalar function stationarity and the quality of the scalar sampling even if its frequency is low. The first criterion is relative to the analyser resolution, which must be better than the difference ΔX of scalar concentrations between the two reservoirs. Then, three quality criteria, defined to control the scalar function stationarity, are applied to the scalar function. This function is measured during the sampling period of the REA by the same low temporal resolution analyser used to quantify the reservoir concentrations. The last criterion, based on the comparison of the mean concentration of reservoirs to the atmospheric one, is aimed at controlling the REA sampling quality.

Before applying the chemical criteria enumerated above, a first sample selection can be made simply based on the mean atmospheric concentration. In fact, high mean concentrations can be associated with polluted plumes, which certainly do not fulfil homogeneity and stationarity hypotheses.

The aim of the following section is to propose a methodology to analyse the quality of the chemical function. It is obvious that the application of these quality criteria strongly depends on the chemical compound measured and the analyser used. Here, the application of the chemical criteria to NO_x functions reveals some limits of the REA sampling system used.

3.3.1. *Analyser Resolution Criterion*

The first criterion of this group is related to the technical characteristics of the analyser used for the scalar measurements. Samples for which the difference in scalar concentrations between the two reservoirs is lower than the analyser resolution are flagged. A REA user should be very careful with these samples representing very small or zero fluxes, and there is the need to investigate whether these fluxes are really zero (for instance at nighttime) or result from a measurement error. In the first case, the measurement is not erroneous but it is unreliable since ΔX is less than the analyzer resolution. This resolution is 0.05 ppb for the NO_x analyser used in this REA system. The concentration difference between the two reservoirs does not present a marked diurnal variation (Figure 5a), with very scattered values during the night. It is always positive between 0800 UTC and 1800 UTC with a mean maximum value of 0.5 ppb at noon.

3.3.2. *Scalar Linear Drift Criterion*

Existence of a linear drift in the scalar function is the first indicator of a non-homogeneous concentration. The linear regression line (least squares best-fitted line) of the temporal evolution of the scalar during the sampling sequence is calculated. If the scalar function is homogeneous, the fitted line should be horizontal (no drift). Thus, deviation from the horizontal, as measured by the slope of the regression line, is used as a criterion to check the presence of a linear drift in the scalar function. Figure 6 shows a chemical function and its linear drift. The hatched curve illustrates the modifications introduced over the function when the linear drift is removed. The vertical and horizontal hatched areas indicate where the chemical concentration is respectively increased and decreased when the linear drift is removed. It should be noted that the presence of a linear drift in the scalar function during the sampling period, coupled with a non-homogeneous and non-equidistributed sampling (expressed by the CRS_{-t+} criterion defined above), may strongly affect the ΔX value. Figure 5b illustrates the mean diurnal variation of the linear drift of the

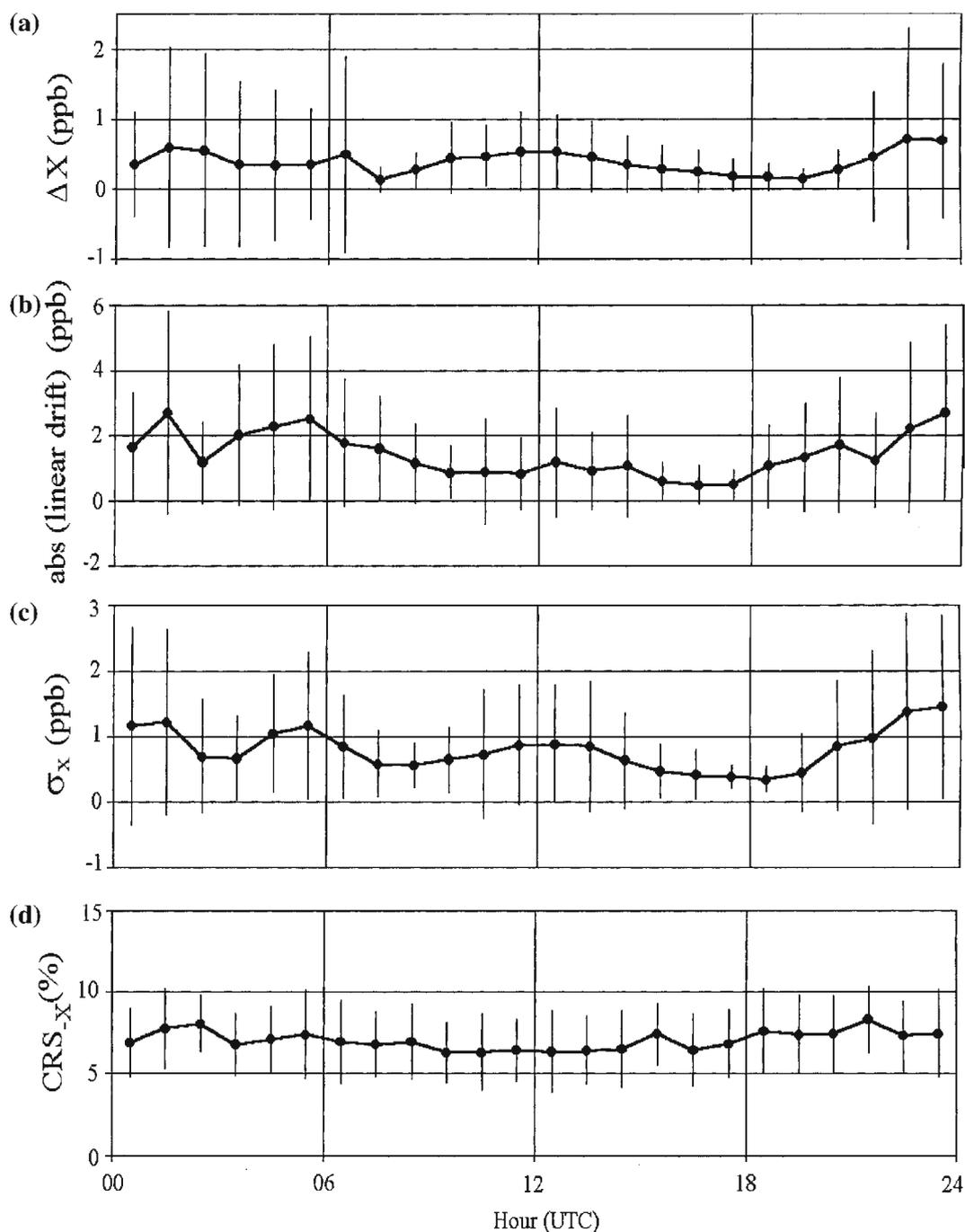


Figure 5. Mean diurnal variation of criteria related to the chemical scalar, for the “Maize” site: (a) concentration difference between the two reservoirs; (b) linear drift of the mean atmospheric concentration; (c) standard deviation, and (d) CRS_{-x} . Vertical bars represent the standard deviation around the mean value indicated by the solid line.

NO_x concentration expressed in absolute values for the “Maize” experiment. The linear drift is, on average, around 1 ppb during the day and 2.5 ppb during the night, but it can reach 5 or 6 ppb if the important scattering of individual values of this criterion during the night is taken into account.

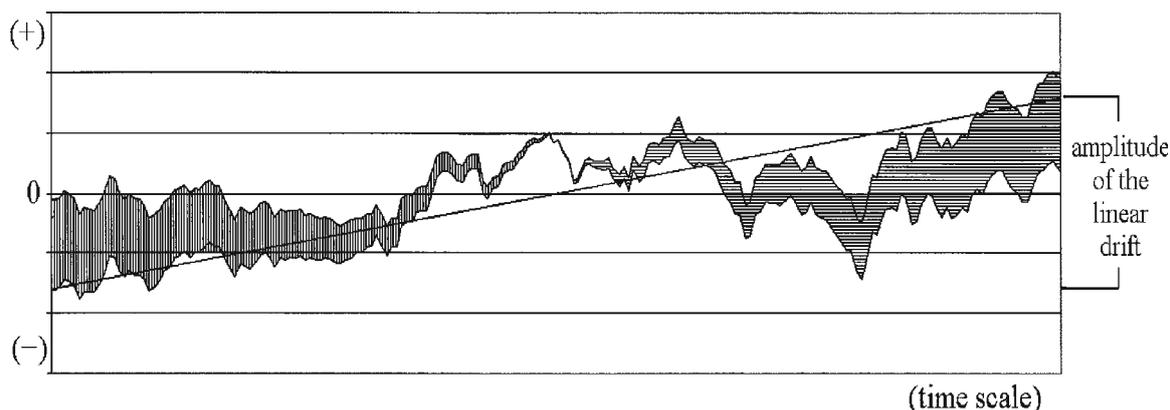


Figure 6. Illustration of the scalar function, its linear drift estimated by the least squares method and the detrended function. Vertical and horizontal hatched areas indicate where the chemical concentrations are respectively increased and decreased when the linear drift is removed.

3.3.3. Standard Deviation Criterion

The standard deviation of the chemical compound function was calculated after removing the linear drift. While the linear drift shows the non-stationarities over a time scale close to the sampling time duration, the standard deviation allows checking for the presence of heterogeneities at a shorter time scale.

The mean diurnal variation of the standard deviation criterion is almost similar to that of the linear drift, with the lower values (0.3 up to 0.9 ppb) during the day (Figure 5c). According to this criterion, the most suitable conditions to measure NO_x fluxes seem to occur around 1830 UTC.

Delon (1999) and Delon et al. (2000) showed that the EC and REA methods are similarly affected by the existence of low frequency drifts in the w or scalar function.

3.3.4. CRS_{-x} Criterion

The concept of this criterion has been previously developed, in the section presenting the ‘Dynamic’ criteria (3.1.3). It characterises the stationarity and the homogeneity of a sample. The CRS_{-x} calculated for the NO_x functions is almost constant with time and on average equal to 7% (Figure 5d). It is consistently higher than CRS_{-w} .

3.3.5. Concentration Centrality Criterion

Theoretically, the average of the concentrations of the two reservoirs, $\bar{X}_{\text{reservoirs}} = (\bar{X}^+ + \bar{X}^-)/2$, should be close to the mean atmospheric concentration, \bar{X}_{atm} , measured during the sampling period, or at the very least, the mean atmospheric concentration value should fall between \bar{X}^+ and \bar{X}^- . Comparison between these two mean concentrations (reservoirs and atmosphere) can be employed as a scalar measurement quality criterion. This

criterion, denoted $\Delta\bar{X}_{\text{atm}}$, is defined as:

$$\Delta\bar{X}_{\text{atm}} = 100 \frac{2|\bar{X}_{\text{atm}} - \bar{X}_{\text{reservoirs}}|}{|\bar{X}^+ - \bar{X}^-|}, \quad (7)$$

where \bar{X}_{atm} lies outside the $|\bar{X}^+ - \bar{X}^-|$ range when this ratio is greater than 100%. As a consequence, measurements can be considered of good quality when the value of this criterion is lower than 100%. Unfortunately for the set of data used in this study, $\Delta\bar{X}_{\text{atm}}$ values, ranging on average between 100 and 250%, usually exceed this cut-off limit. The problem lies in the different pathways followed by the flow during the reservoirs and atmospheric concentration measurements. The pressure drop might have been different in the two different (direct and reservoirs) measurement circuits, implying a different pressure in the analyser cell. However, this malfunction does not affect ΔX and the flux estimate, since the air flow remains constant during the sampling and analysis of the negative and positive reservoirs, which have symmetric circuits.

4. Results of the Criteria Application to Experimental Data

The next sections present the results derived from the application of the criteria defined above. They are applied to the data collected at the two experimental sites, ‘‘La Crau’’ and ‘‘Maize’’.

4.1. CUT-OFF LIMIT VALUES AND FLAG RATES OF CRITERIA

Once the criteria have been established, their values are computed for each sample acquired during the two experiments. The next step is to define the upper and lower cut-off limits of criteria used to flag questionable samples. Turbulence structure, selection algorithms and scalar characteristics are not optimal for these samples. For a given site, definition of these limits is made by an empirical method, through the distributions of the computed criteria values for the whole campaign. When these distributions are symmetric, cut-off limit criteria correspond, more or less, to $\pm 3\sigma$ around the mean value. Practically, samples with values situated in the tails of the distributions, outside the area of $\pm 3\sigma$, are flagged. For a non-symmetric distribution, criteria limits are properly adjusted with an iterative method until the skewness of the distribution reaches a value close to one. Table I presents the cut-off limits and percentages of flagged samples for ‘Dynamic’ and ‘REA operation’ criteria for both sites. Table II focuses on the application of ‘Chemical Scalar’ criteria to data collected at the ‘‘Maize’’ site.

TABLE I

Presentation of the low and high cut-off limits of “dynamic” and “REA functioning” criteria defined in paragraph 3, for ‘La Crau’ and ‘Maize’ sites.

Criteria	Skew	Kurt	L_ε (m)	T_ε (s)	CRS-W	$\sigma_w/U(\times 10^2)$	$t+$ (s)	CRS- $tT+$	Total (%)	Net (%)
‘La Crau’ site										
Low cut-off limit	-0.40	-	-	-	-	3.00	0.57	-		
High cut-off limit	1.00	6.00	20.00	8.00	8.00	13.00	1.39	6.00		
% Flag samples	3	3	1	6	4	9	4	3	33	15
‘Maize’ site										
Low cut-off limit	-0.50	-	-	-	-	3.00	0.52	-		
High cut-off limit	1.00	8.00	16.00	100.00	14.00	22.00	1.65	6.00		
% Flag samples	4	3	3	5	1	11	3	8	38	23

The percentage of flagged samples by each criterion is also presented. The column entitled “Total (%)” is obtained by summing all the percentages of flagged samples. Then, “Total (%)” value is the percentage of flagged samples that should have been obtained if all samples have been flagged by only one criterion. As samples are flagged by several criteria simultaneously, the real percentage of flagged samples is always lower than the previous one (Total) and is presented in the last column titled “Net (%)”

TABLE II

Presentation for ‘Maize field’ site of the low and high cut-off limits of “Chemical” criteria defined in paragraph 3.

Criteria	Analyser resolution ΔX (ppb)	Std deviation/ ΔX (%)	Linear drift/ ΔX (%)	CRS- NO_x (%)
‘Maize’ site				
Cut-off low limit	-	-	-	-
Cut-off high limit	0.05	10.2	20	16
% Flag samples	16	10.9	12.4	7.0

The percentage of flagged samples by each criterion is also presented.

In most cases, cut-off limits given in Table I are not similar for the two ecosystems. These results show that the limits chosen are not universal and have to be adapted according to the site studied. The percentages of the samples flagged by each criterion, which allow the evaluation of criteria strictness, are different for the two sites. As an example, although the

turbulence intensity criterion has a higher cut-off limit for the “Maize” site (0.22) than for “La Crau” (0.13), it flags 11% for “Maize” versus 9% for “La Crau”. For both sites, turbulence intensity (σ_w/u) is the most severe criterion as it flags more samples than the others do. Table I also shows that the flag rate of the dissipation criteria depends on which unit is used (seconds or metres). It flags more samples when it is expressed in seconds than in metres. This information is important since it reveals that high values of T_ε associated with light winds are flagged, whereas the corresponding values of L_ε are not flagged. Moreover, although the ratio of cut-off values $T_{\varepsilon_Maize}/T_{\varepsilon_Crau}$ is of the order of 2.5, the $L_{\varepsilon_Maize}/L_{\varepsilon_Crau}$ ratio is reversed, and equal to 0.8. This implies different wind conditions for the two sites, with much lighter mean horizontal winds in the maize field than in the “La Crau” plain.

Concerning the ‘Chemical Scalar’ criteria (Table II), ‘Analyser resolution’ is the most severe of the criteria since it flags 16% of the samples. This means that 16% of the samples display a concentration difference between updraft and downdraft reservoirs which is lower than the analyser resolution. In Table II, ‘Standard deviation’ and ‘Linear drift’ criteria are normalised by ΔX in order to point out the very high value of the cut-off limit, which is about 10 ΔX for the ‘Standard deviation’ criterion, and 20 ΔX for the ‘Linear drift’ one. In spite of these high cut-off limits, these two criteria flag only 10 and 12% of the samples. Finally, the most lenient ‘Chemical Scalar’ criterion is the CRS_{-x} .

The column entitled ‘Total (%)’ in Table I is obtained by adding the percentages of flagged samples by each criterion, and represents the total percentage of the flagged samples for the whole set of criteria. In fact, since samples are simultaneously flagged by several criteria, the true percentage of the flagged samples presented in the last column (Net (%)) is always lower than the former (Total). This implies that when the difference between ‘Total’ and ‘Net’ percentages is large, an important number of samples is flagged by more than one criterion. The ‘Total’ percentage is 33% for “La Crau” site and 38% for “Maize” site, and the ‘Net’ percentage is 15% and 23% for “La Crau” and “Maize” site respectively. This shows that there are overlapping criteria. Although the cut-off limits of criteria are defined in an empirical way, criteria overlapping could be used as a mean of ensuring that a sample should be flagged.

Flag rates of “Dynamic” and “REA operation” criteria exhibit a clear diurnal variation (Figure 7). Actually, criteria are more severe during the night as the majority of the flagged samples are located in the nocturnal period. Two secondary maximum (0700 and 1900 UTC) which correspond to the sunrise and sunset transition periods, are revealed. These maxima are probably related to an increase of the non-stationarity of w functions at that time. Except for the ‘Resolution analyser’ criterion, flag

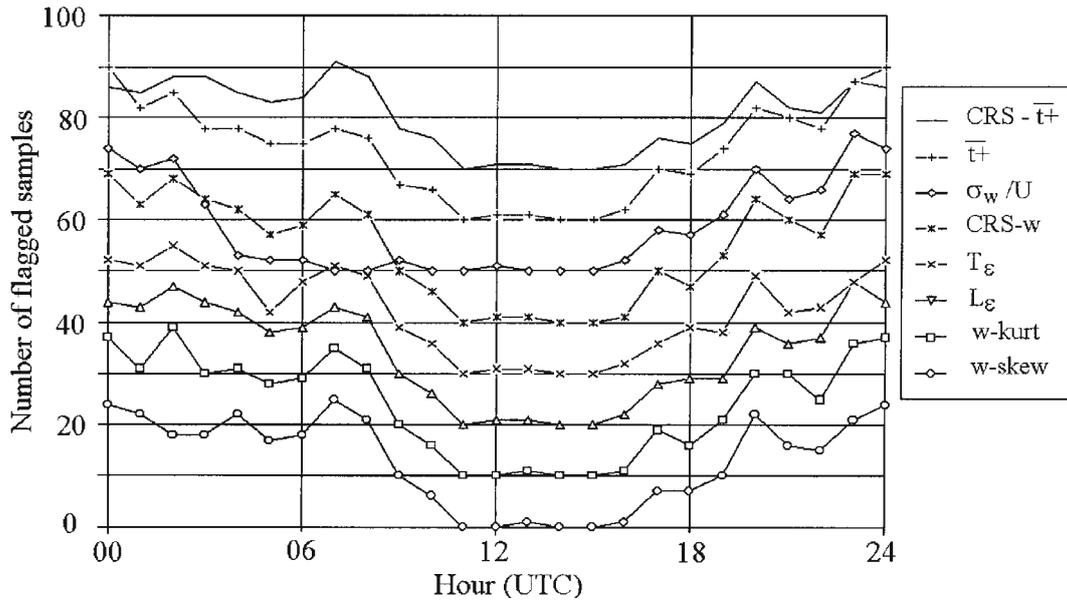


Figure 7. Diurnal variation of the rejection rate, for each one of the nine ‘Dynamic’ and ‘REA operation’ criteria, for the “Maize” site. The x -axis represents the hour of day (UTC), and the y -axis represents the number of flagged samples. In order to represent all these curves in the same graph, each curve is shifted by ten units.

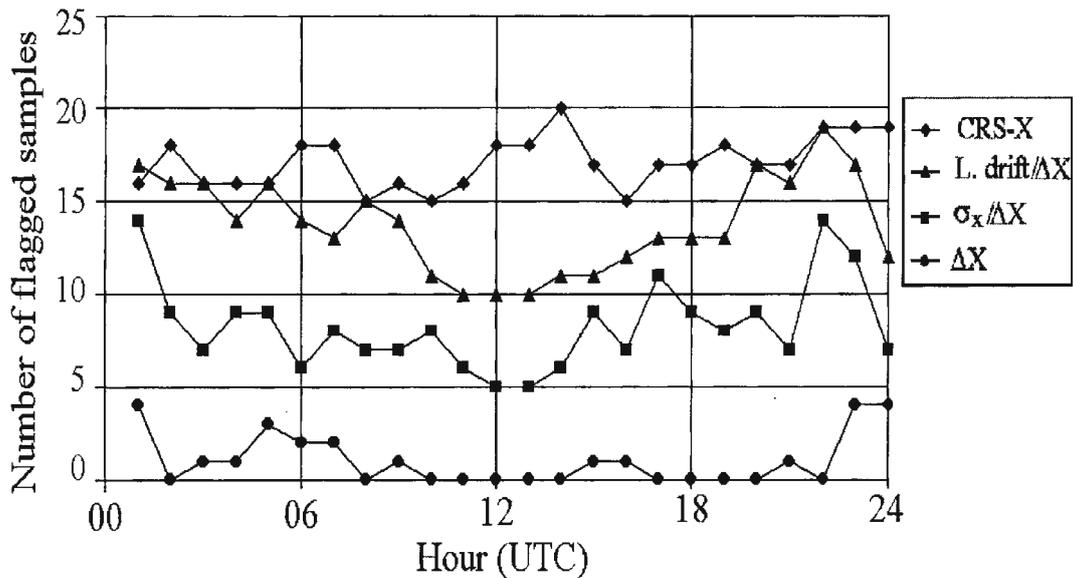


Figure 8. Diurnal variation of the rejection rate of scalar criteria, for the “Maize” site. The x -axis represents the hour of day (UTC), and the y -axis represents the number of flagged samples. Each curve is shifted by five units.

rates of ‘Chemical Scalar’ criteria (Figure 8) have no pronounced diurnal variation. However, the total number of flagged samples for ‘Dynamic’ and ‘REA operation’ criteria (Figure 9a), and for ‘Chemical Scalar’ criteria’ (Figure 9b), show a similar daily evolution. Daytime samples are less flagged than the nocturnal ones.

Some differences can be noticed however. First, the ‘Dynamic’ and ‘REA operation’ criteria flag curve is not symmetric since its minimum

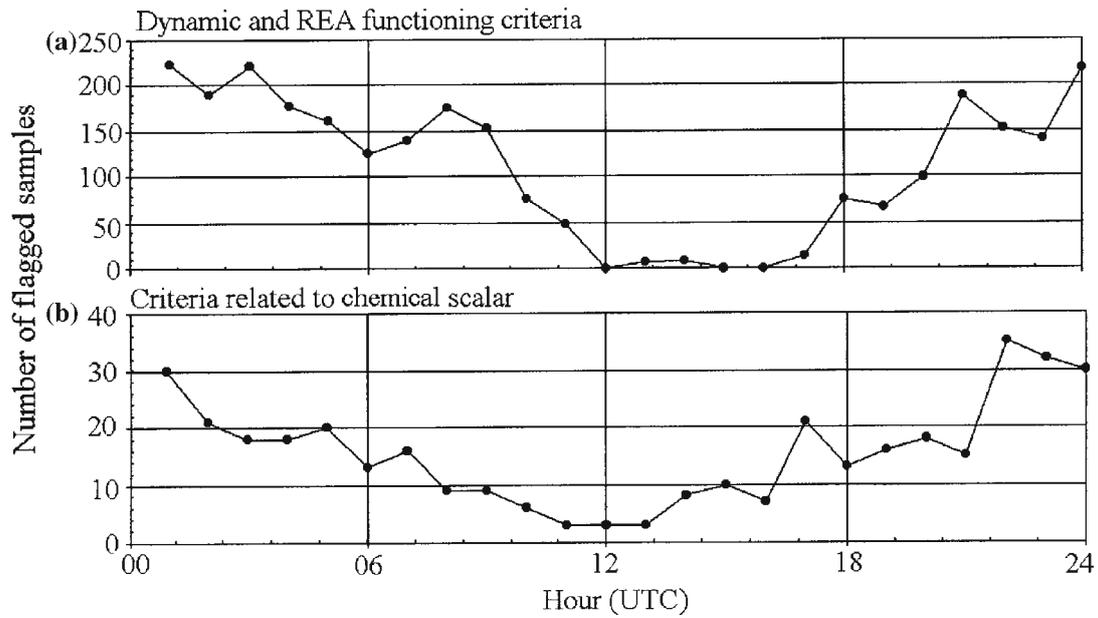


Figure 9. Diurnal variation of the flagged rate for the ensemble of (a) ‘Dynamic’ and ‘REA operation’ criteria; and (b) ‘Chemical Scalar’ criteria.

occurs between 1200 and 1700 UTC. Samples collected during the ABL establishment exhibit a high flag rate. Secondly, the ‘Chemical Scalar’ curve is symmetric around noon but contrary to the other criteria, never reaches zero.

4.2. FLAG RATE OF “DYNAMIC” AND “REA OPERATION” CRITERIA ACCORDING TO MEAN DYNAMIC AND STABILITY CONDITIONS FOR THE “MAIZE” SITE

In this section, the study focuses on the strictness of the ‘Dynamic’ and ‘REA operation’ criteria obtained for the “Maize” site. An analysis of the

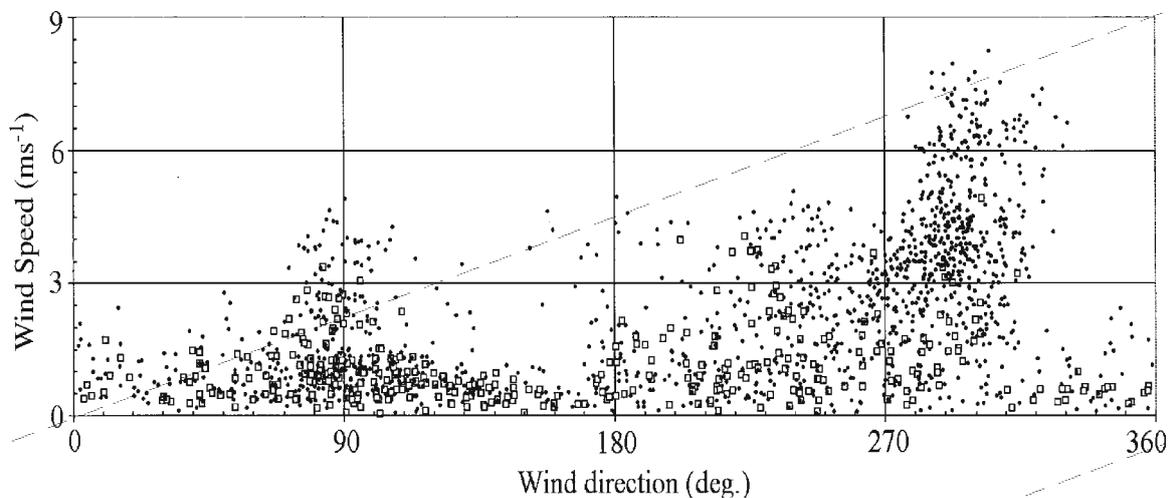


Figure 10. Individual samples as a function of the wind speed in m s^{-1} (y-axis) and wind direction in degrees (x-axis). Flagged samples are represented by large open squares.

flagged samples as a function of the mean dynamic conditions (wind speed and direction) is presented in Figure 10. Strong winds (mean speed $> 4 \text{ m s}^{-1}$) appear for two directions: east and north-west. Flagged samples (open squares) seem to be insensitive to wind direction but they are always associated with light winds ($< 4 \text{ m s}^{-1}$), implying again inhomogeneous and non-stationary w functions.

To interpret these findings, the conditions of atmospheric stability corresponding to flagged samples are examined. Atmospheric stability is expressed through the surface boundary-layer scales of wind velocity (u_*) and temperature (T_*). Distribution of the flagged samples is presented in a bi-dimensional graph as a function of the scales u_* and T_* (Figure 11). In that figure, flagged samples are marked with open coloured squares, whereas small dots are used for the non-flagged ($u_* - T_*$) pairs. The colour code indicates their time position in the diurnal cycle, with cold and hot colours for night and day, respectively. From Figure 11, one can see that most of the flagged samples correspond to stable and neutral conditions, mainly appearing during the nocturnal period. This result confirms the previous conclusions deduced from Figures 7 and 9a. However, a number of the flagged samples corresponds to conditions of high convective instability (T_* varying from -0.6 down to -1.6 K with u_* lower than 0.3 m s^{-1}). This group of samples is related to the morning flag maximum well illustrated in Figure 9a. This daytime period coincides with the increase of the solar radiation, which is the key factor contributing to the boundary-layer

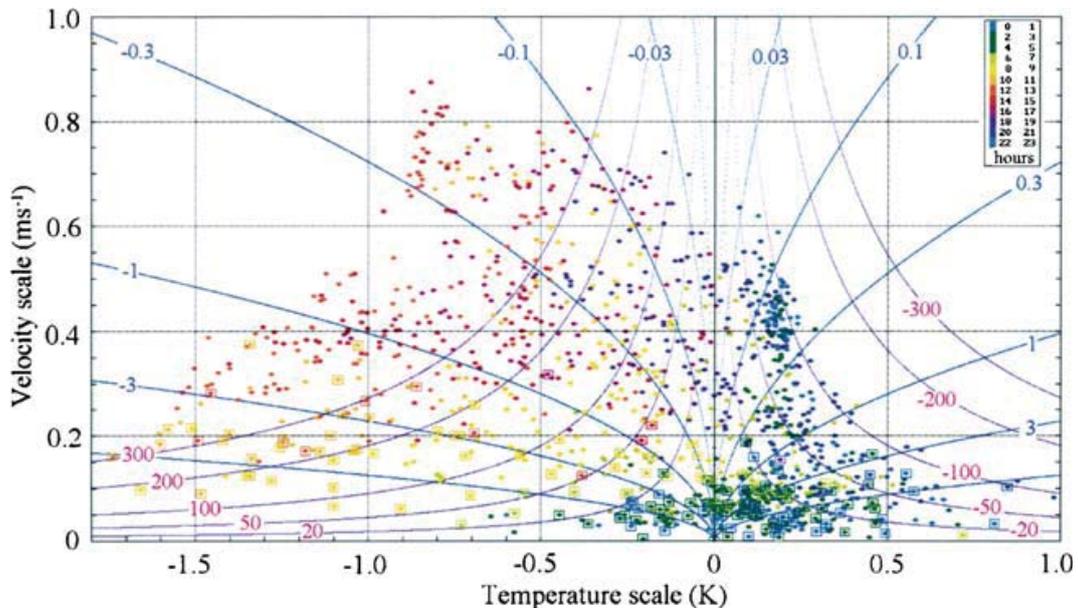


Figure 11. Samples as a function of the atmospheric stability for the “Maize” site. x-axis: surface boundary layer temperature scale (T_*), y-axis: friction velocity (u_*). Hyperbolic and parabolic lines represent isovalues of sensible heat fluxes (W m^{-2}) and isovalues of stability conditions, respectively. Small coloured dots correspond to $u_* - T_*$ couples. Open coloured squares correspond to flagged samples. Colour code indicates the time in UTC.

development. The high convective instability conditions established at that time result in a deviation of the turbulent functions from the Gaussian distribution. Indeed, for the period between 0600 and 1000 UTC, skewness and kurtosis are the most severe among the whole set of criteria (Figure 7). Moreover, free convection implies the formation of large-scale eddies, which, in their turn, make turbulent functions non-stationary. This conclusion is confirmed in Figure 7 by the relatively high values of the CRS_w and the turbulence intensity at this period of the day.

Previous results confirm that atmospheric stability conditions have an impact on data quality. Indeed, depending on what the stability conditions are, necessary assumptions required for the flux calculation are more or less validated. ‘Dynamic’ criteria directly guarantee the measurement quality as they are derived from the turbulent function of the vertical wind velocity. However, it is found that the REA functioning is also affected by the

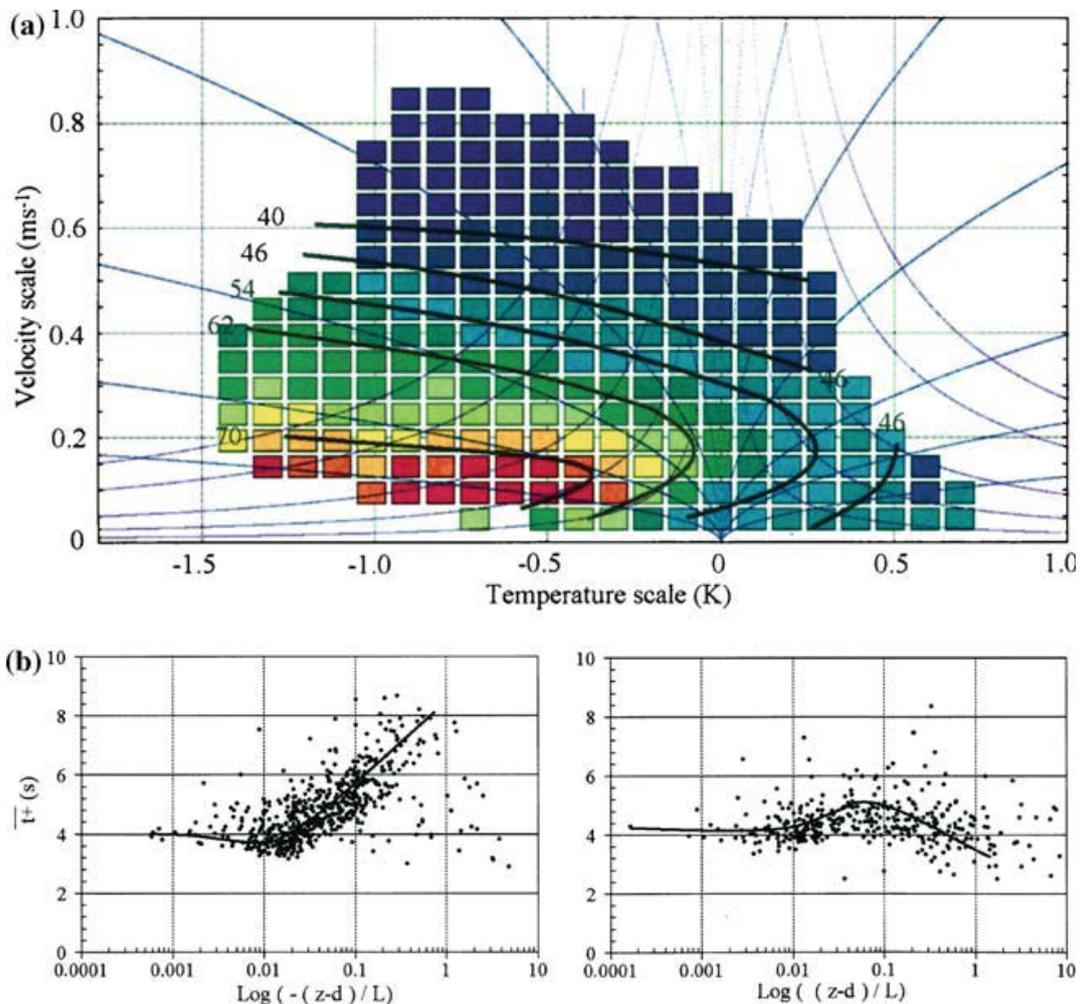


Figure 12. (a) Bi-dimensional graph of mean positive selection time in seconds (multiplied by 50) as a function of u_* and T_* . The same representation as in the Figure 11 is used. (b) The dependence of the mean duration of positive selection (t_+) on the atmospheric stability expressed by $(z-d)/L$ parameter: left for unstable ($(z-d)/L < 0$) conditions and right for stable ($(z-d)/L > 0$) conditions.

atmospheric stability conditions. To illustrate this remark, a ‘REA operation’ parameter that is also used as a criterion, the mean time of positive selection (t_+), is presented in a bi-dimensional graph as a function of u_* and T_* (Figure 12a). For unstable conditions, the highest values of t_+ are associated with low dynamic instability ($u_* < 0.2$) and t_+ decreases with increasing u_* . In the case of stable conditions ($T_* > 0$), t_+ has a weaker variation range. This type of representation shows the dependency between the studied parameter (here t_+), and the surface-layer stability given by the two scales u_* and T_* . When compared to $(z - d)/L$ isolines, this allows one to appreciate the difference between experimental and ideal representations. Coming back to the classical theory of the surface ABL, one can obtain a simple relation describing the dependence of the parameter (t_+) upon the atmospheric stability. Figure 12b shows the distribution of t_+ as a function of atmospheric stability expressed through the parameter $\zeta = (z - d)/L$, where L is the Obukhov length scale, d the displacement distance and z the measurement height ($(z - d)/L > 0$ for stable conditions and $(z - d)/L < 0$ for unstable conditions). This graph shows that, under neutral conditions, t_+ is relatively constant at about 1 s, which actually is a low value. Under stable conditions, it slightly increases up to 1.3 s and then decreases down to 0.9 s with increasing stability. The strongest variations are observed for unstable conditions since t_+ has a maximum value of about 1.7 s for $(z - d)/L$ close to 1.

5. Conclusions

Reliable flux measurements can be made with the REA method as long as the users undertake a quality control of the measurements and the REA functioning, defined in the theoretical framework of this paper and in the companion paper. Such quality control is essential because the REA behaves like a ‘black box’ since screening of the air sampling procedure in real time is not possible. Furthermore, contrary to the EC method, in the REA case the turbulent (high) frequencies of the scalar function are not measurable. Additionally, checking of the turbulence stationarity and homogeneity is indispensable to ensure the measurement quality.

In this work, using all the available information, an extended methodological approach consisting of 12 criteria is developed for characterising the REA measurement quality. These criteria are classified in three groups: ‘Dynamic criteria’, ‘REA operation criteria’, and ‘Chemical scalar criteria’, based on the dynamic measurements (w and u), on selection algorithm, and on the scalar low frequency properties, respectively.

Criteria are a powerful data quality control tool for the REA users. They allow one to check the fulfilment of the stationarity and homogeneity

assumptions, investigate the application limits specific to the REA sampling and the analysis process, and to identify samples deviating substantially from the optimal measurement conditions leading to questionable flux estimates. Dubious samples are flagged through the procedure of the criteria cut-off limits definition made in an empirical way via the distribution of the computed criteria values and accounting for the specificity of the site.

Apart from the criteria definition, their application to datasets coming from two different experimental sites allowed us to draw a number of conclusions. It showed that the flagged samples are most of the time of poor quality. It also revealed that criteria cut-off limits have to be adapted according to the site studied and the experimental conditions such as the height of measurement. Indeed, they can vary enough between the two sites implying that the data quality is dependant on the site characteristics (underlying surface, roughness, aerological characteristics, local chemistry, ...). More precisely, the set of 'dynamic' and 'REA operation' criteria flags 15 and 23% of samples for the "La Crau" (bare soil) and the "Maize" (vegetation covered) site, respectively. An overlapping of criteria was observed since a sample can be simultaneously flagged by more than one of them.

Values of 'Dynamic' and 'REA operation' criteria are closely related to the stability conditions of the atmospheric surface layer (ASL). The study conducted with the "Maize" data showed that the maximum of samples flagged by 'Dynamic' and 'REA operation' criteria is found under: (i) stable or neutral nocturnal conditions corresponding to weak fluxes, disturbance in the ASL dynamics by stable flow or wave motions and intermittent turbulence, and (ii) highly convective instability regimes occurring early in the morning. Taking into account the diurnal pattern of criteria values and the flag rate behaviour, it is concluded that the daytime period is more favourable for REA measuring than the nighttime. During the daytime, REA functioning is optimal in the evening under dynamically unstable conditions than in the morning.

Contrary to 'Dynamic' and 'REA operation' criteria, which are directly or indirectly associated with the turbulent function (w), the 'Chemical' ones are applied to a low frequency function, exhibiting a different diurnal pattern. They do not display a marked diurnal cycle, being similarly sensitive, as low frequencies induce the same deformation day and night. Their flag rates are almost constant during daytime and relatively independent of the ASL stability evolution.

Having emphasised that the data quality is related to the stability conditions through the vertical and mean wind velocity series, the stationarity of the selection algorithm parameters, the site chemistry through the chemical scalar trends and the site characteristics through the cut-off criteria limits, such an approach seems indispensable for a 'blind' method like the REA.

This work is a first attempt to propose a complete REA measurement quality diagnostic scheme. It can, however, be further developed. For instance, here, criteria for cut-off limits are empirically defined, while as a next step, we could look at limits based on theoretical considerations. They could also be empirically defined based on a database referring to a great variety of fully documented sites and in relation to the ASL state. Such a database is especially useful for criticising non-continuous REA measurements because it could allow an *a priori* knowledge of the cut-off limits and of the selection algorithmic parameters such as the asymmetric threshold on w signal (see part I). Finally, it can be deduced from the significant overlapping in the criteria responses that a more sophisticated selection should be based on a combination of several criteria for each sample.

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