

## **A guide to identifying periods without apparent ebullition**

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In order to fit the empirical ebullition threshold, the user should identify half-hour periods in which ebullition is not present in the high frequency CH<sub>4</sub> concentration data. These periods should be sampled from the set of periods **meeting all quality requirements for both reference scalars**, span as much of the analysis period (e.g., a growing season) as possible, and encompass a variety of times of day and meteorological conditions (e.g., friction velocity, stability). Because the ebullition threshold width is a function of CH<sub>4</sub> standard deviation ( $\sigma_m$ ), the selected non-ebullitive periods should also encompass the range of  $\sigma_m$  observed in the set of best quality periods. Follow the guidelines below to select non-ebullitive periods for your site-year of data.

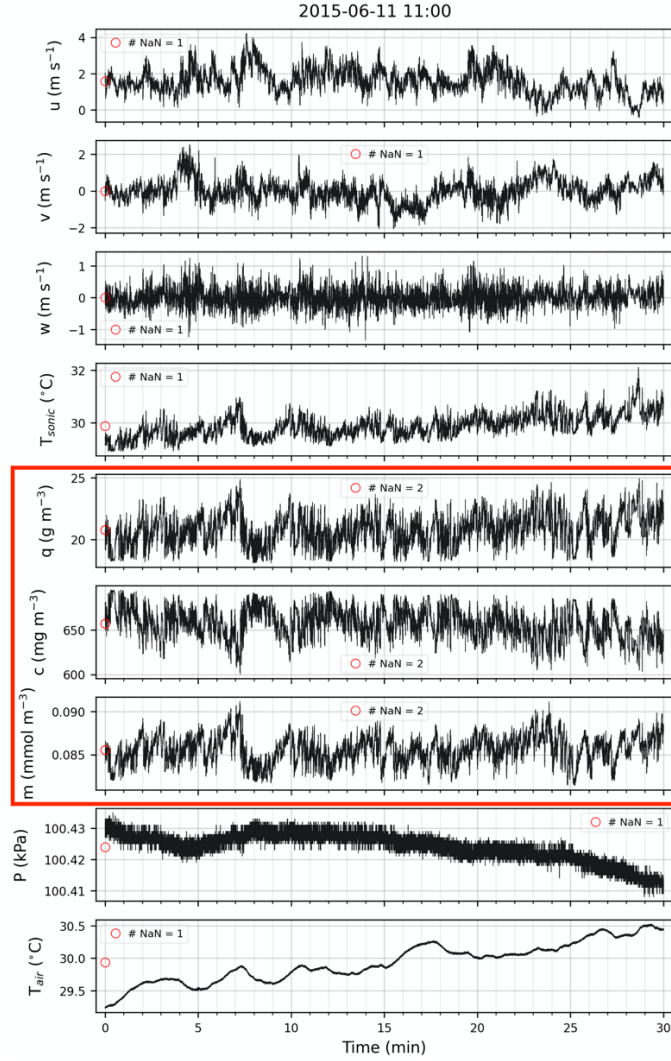
### **Selection guidelines**

Ebullition appears in the CH<sub>4</sub> concentration time series as sharp, intermittent spikes which are unique to the CH<sub>4</sub> data and not present in the other reference scalar time series. Over the course of any half-hour observation period, time series of each gas concentration are generally characterized by ramp-like structures which are indicative of diffusive exchange. Ebullition may present itself as intermittent spikes uniquely deviating from the ramp-like structures in the CH<sub>4</sub> time series, or in some cases where ebullition is particularly prominent, the magnitude of these intermittent spikes may be so large as to make the individual ramps hard to distinguish. In either of these cases, the period in question would be considered as having non-negligible ebullition and thus excluded from further consideration as a non-ebullitive period. Note that periods in which any of the gas concentration time series has significant trends should be omitted from consideration as a non-ebullitive period; these trends typically cause divergent RMSD– $\sigma_m$  relationships from periods without trends (i.e., RMSD and/or  $\sigma_m$  magnitude are impacted by the trends). The user should aim to identify approximately at least 5–8 non-ebullitive periods per month of eddy covariance data.

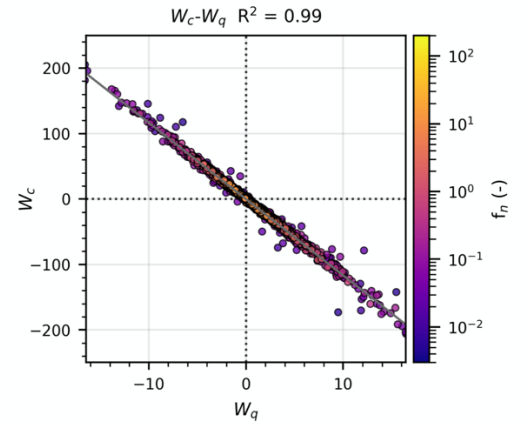
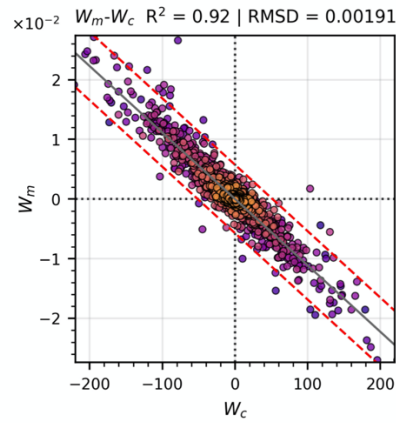
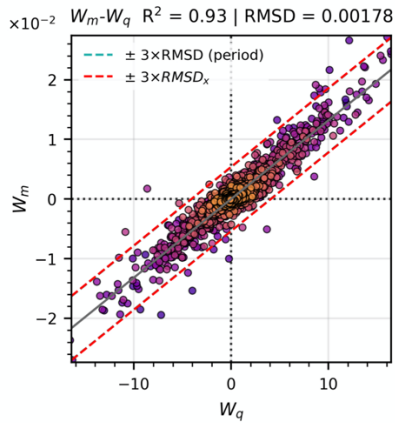
After identifying a selection of non-ebullitive periods, the user should make and examine wavelet coefficient ( $W_x$ ) scatterplots for further confirmation that the selected non-ebullitive periods are appropriate. This set of scatterplots should include the following set of relationships: methane wavelet coefficients ( $W_m$ , y-axis) versus reference scalar wavelet coefficients ( $W_q$  for H<sub>2</sub>O,  $W_c$  for CO<sub>2</sub>, x axis; one plot for each reference scalar) and, if using more than one reference scalar, plot the wavelet coefficients of each reference scalar against one another. Critically, the color of the wavelet coefficients should correspond to their respective normalized frequency values so that the user can see if deviations from the line of best fit are associated with low or high frequency fluctuations. To confirm the period for inclusion in the set of non-ebullitive periods, the methane–reference scalar plots should show coefficients generally clustered around the line of best fit, with few (or no) high frequency coefficients falling far away from this line. When using multiple reference scalars, it is critical to also ensure that the reference scalar wavelet coefficients have *very strong* scalar similarity to preclude events in the reference scalar time series from driving the detected ebullition events.

## Examples of non-ebullitive and ebullitive periods

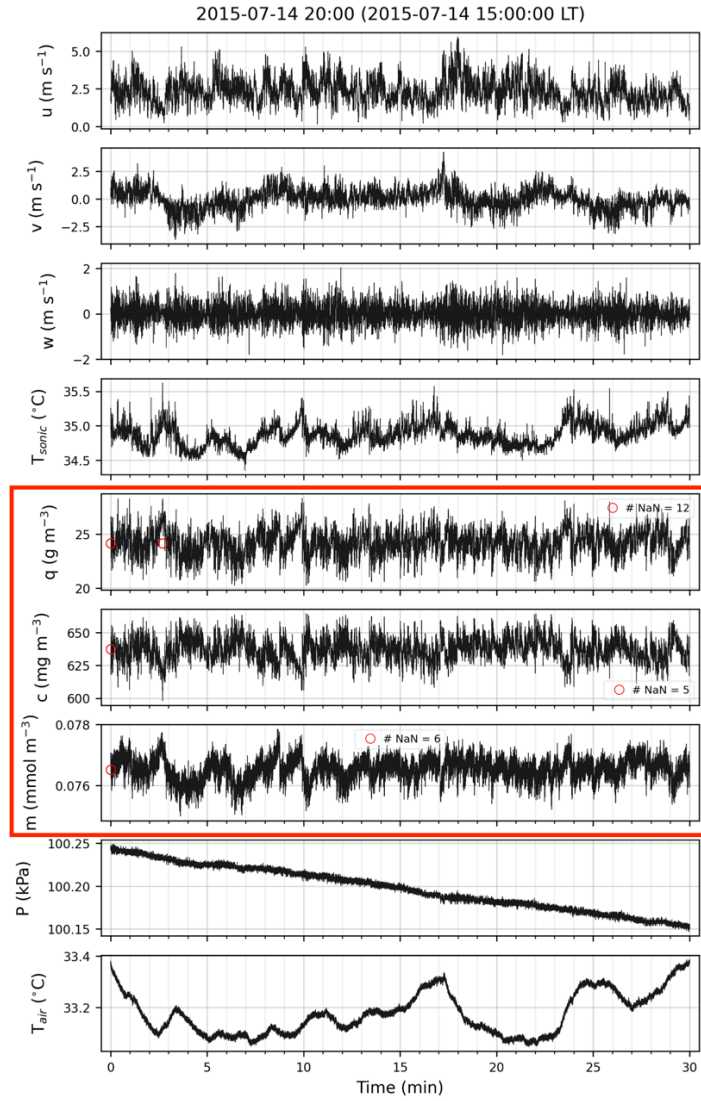
- Non-ebullitive (example 1):



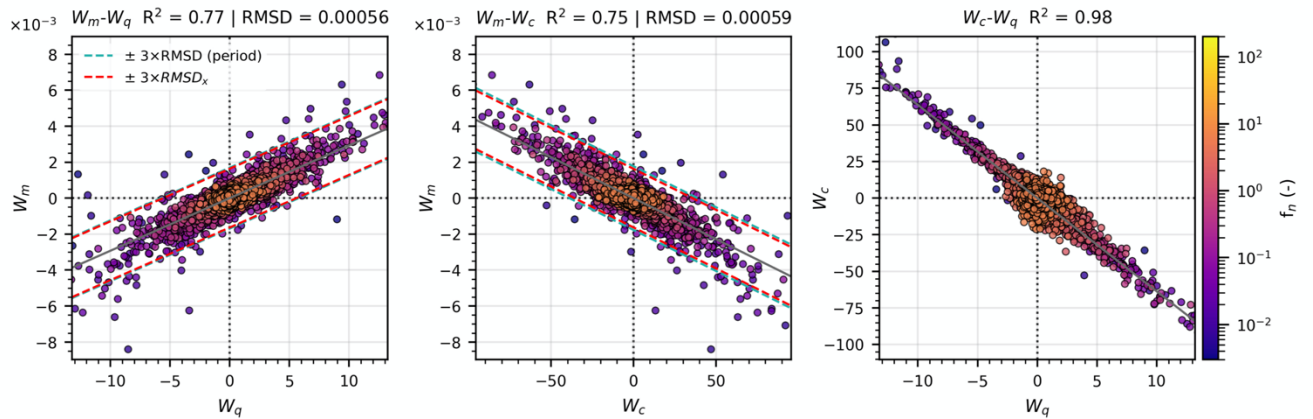
Way3-Summer-2015 11-Jun 11:00  
 $F_{CH_4, tot} = 0.21 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $LE = 371 \text{ W m}^{-2}$  |  $F_{CO_2} = -41.4 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $H = 22 \text{ W m}^{-2}$  |  $u_* = 0.24 \text{ m s}^{-1}$   
 $\sigma_m = 0.0016 \text{ mmol m}^{-3}$  |  $\bar{u} = 1.58 \text{ m/s}$  |  $T_{air} = 29.9 \text{ }^\circ\text{C}$  |  $RH = 69 \%$  | wind direction =  $210^\circ$  |  $\frac{z-d}{L} = -0.03$



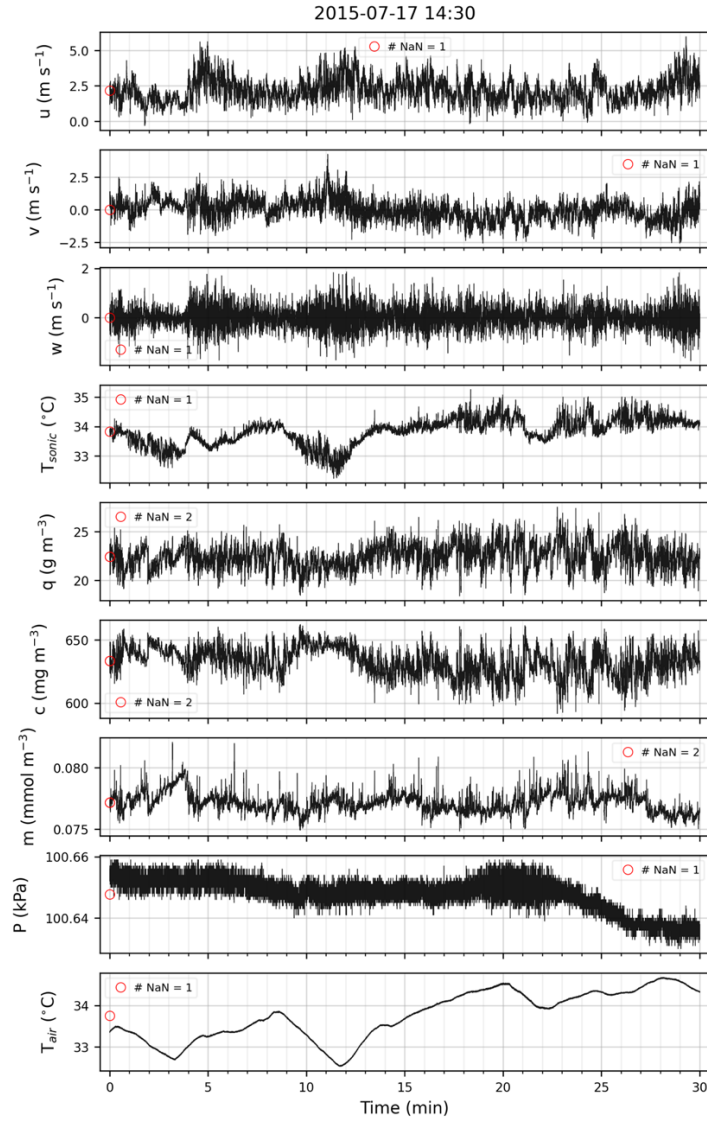
- Non-ebullitive (example 2):



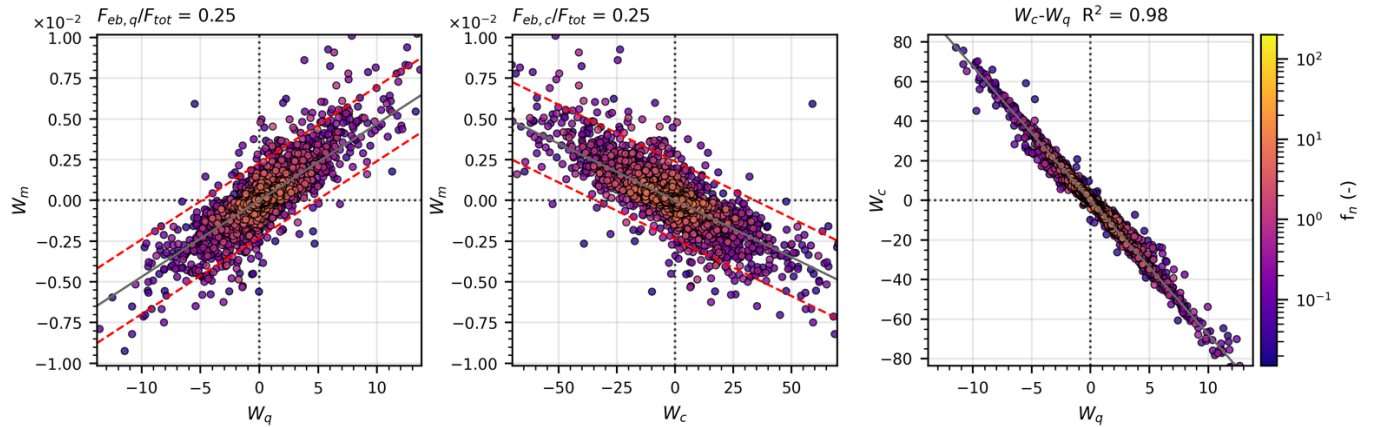
Way4-Summer-2015 14-Jul 15:00  
 $F_{CH_4, tot} = 0.06 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $LE = 479 \text{ W m}^{-2}$  |  $F_{CO_2} = -28.8 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $H = -10 \text{ W m}^{-2}$  |  $u_* = 0.31 \text{ m s}^{-1}$   
 $\sigma_m = 0.0004 \text{ mmol m}^{-3}$  |  $\bar{u} = 2.35 \text{ m/s}$  |  $T_{air} = 33.2 \text{ }^\circ\text{C}$  |  $RH = 67 \%$  | wind direction =  $235^\circ$  |  $\frac{z-d}{L} = 0.01$



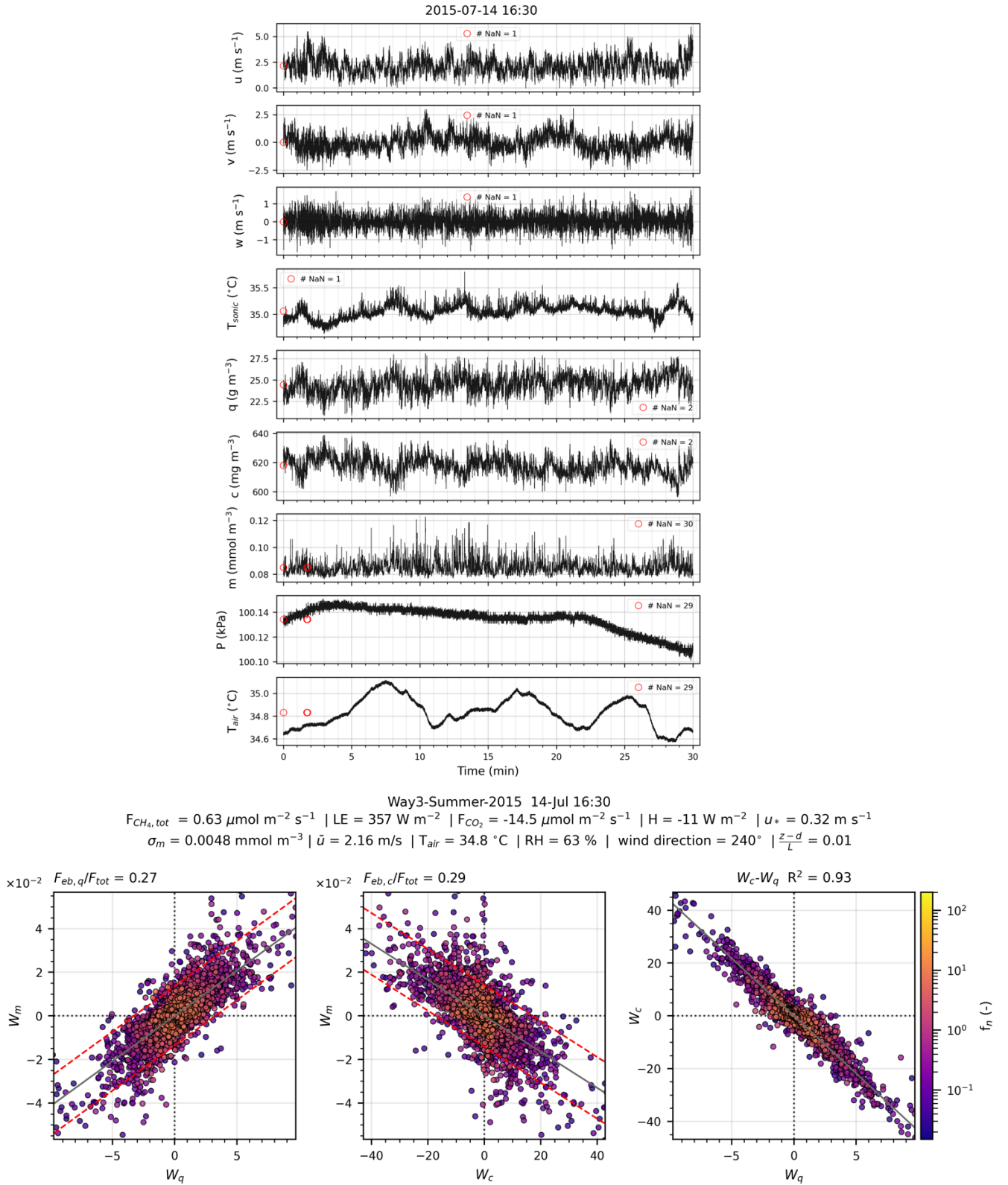
- Ebullitive (example 1, ebullition superimposed on ramp structures):



Way3-Summer-2015 17-Jul 14:30  
 $F_{CH_4, tot} = 0.08 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $LE = 423 \text{ W m}^{-2}$  |  $F_{CO_2} = -26.1 \mu\text{mol m}^{-2} \text{s}^{-1}$  |  $H = -7 \text{ W m}^{-2}$  |  $u_* = 0.30 \text{ m s}^{-1}$   
 $\sigma_m = 0.0008 \text{ mmol m}^{-3}$  |  $\bar{u} = 2.17 \text{ m/s}$  |  $T_{air} = 33.8 \text{ }^\circ\text{C}$  |  $RH = 61 \%$  | wind direction =  $227^\circ$  |  $\frac{z-d}{L} = 0.00$



- Ebullitive (example 2, magnitude of intermittent  $\text{CH}_4$  spikes much greater than ramp amplitude):



### Recommended workflow

1. From the data frame containing periods that meet all quality requirements for both reference scalars, isolate periods in which both  $W_m-W_q$  and  $W_m-W_c$   $R^2$  values (from iteratively reweighted least squares linear regression) are highest (e.g., 0.9 or higher, may need to relax depending on your dataset), as these periods have generally the strongest scalar similarity in the dataset and thus are likely to be less affected by ebullition. Go through the time series plots for each of these periods one by one to check for the presence of apparent ebullition.
2. If more non-ebullitive periods are needed after this first round of screening, consider relaxing the  $W_m-W_x$   $R^2$  threshold criterion and repeat the process for periods not examined in step 1.
3. Plot the distribution of  $\sigma_m$  from all best quality periods, and on the same graph plot the distribution of  $\sigma_m$  from the selected non-ebullitive periods. If the range of  $\sigma_m$  in the non-ebullitive periods generally covers that of the full dataset, proceed to ebullition threshold fitting with these periods. Otherwise, identify the range of  $\sigma_m$  magnitudes in which there are no currently identified non-ebullitive periods. Filter the best quality data frame based on the desired range of  $\sigma_m$ , and continue the screening process.