THESIS

FALCON: A NEW APPROACH TO PROCESSING FLUXES OF AEROSOLS

Submitted by

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ABSTRACT

FALCON: A NEW APPROACH TO PROCESSING FLUXES OF AEROSOLS

The removal processes for atmospheric aerosols impact the radiative balance of Earth and how nutrients and pollutants move. Dry deposition is poorly modeled globally and over many types of surfaces compared to wet deposition. Important surface types on Earth, like the ocean and cryosphere, can be better understood by generating larger datasets of dry deposition measurements over those surface types. Aerosol properties, such as size, influence deposition in addition to surface and meteorological properties. More size-dependent dry deposition measurements can help models improve their dry deposition estimates, especially if those measurements have smaller uncertainties.

Dry deposition velocities can be calculated from vertical aerosol fluxes. Fluxes of aerosols are generally noisier than gas or energy fluxes but can still be found with the eddy covariance technique when fluxes are measured over a long period of time. Using the eddy covariance technique to find aerosol fluxes requires fast (10Hz) measurements using two adjacent instruments. Aerosol concentrations are collected by a portable optical particle spectrometer (POPS) and wind velocities are found with a sonic anemometer. We demonstrate one way of calculating vertical aerosol fluxes over various surfaces. EddyPro and custom scripts process anemometer and POPS data into long-term and continuous flux datasets. This document provides instructions on each step of processing data, starting from the raw files produced and ending at a point where fluxes and deposition velocities are calculated.

TABLE OF CONTENTS

CHAPTER 1 – LITERATURE EVALUATION

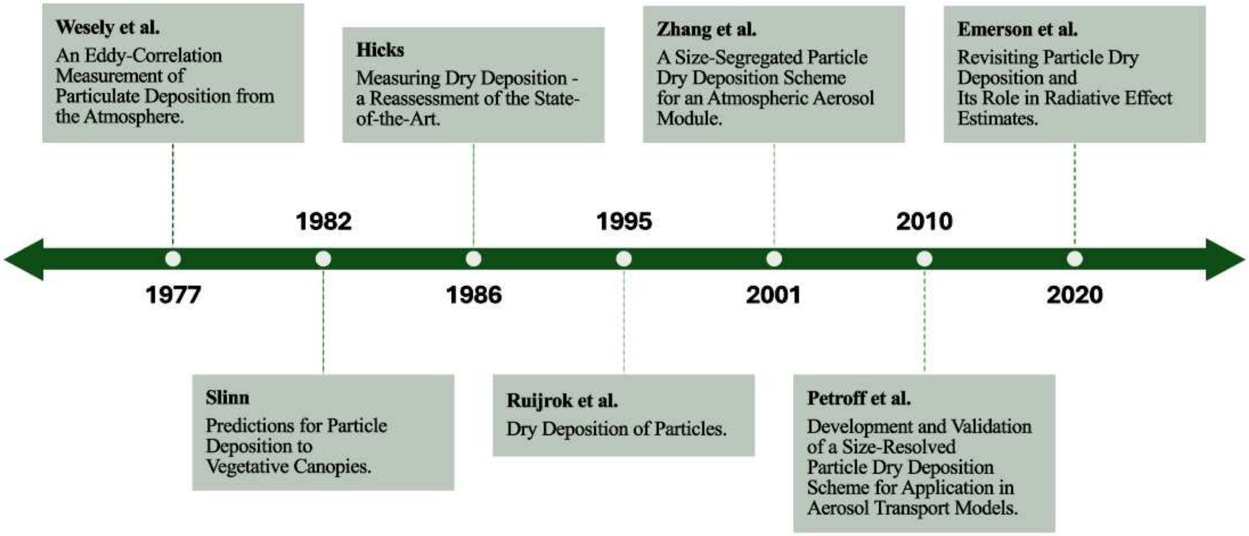

Figure 1. Literature timeline from 1977 to 2020.

Aerosols in the atmosphere impact the radiative balance of Earth and are important to investigate to improve our understanding of human caused climate change. Aerosols affect radiative balance by direct and indirect effects. Aerosol direct effects include aerosols absorbing or scattering radiation in the air, indirect aerosol effects involve aerosols acting as sites for nucleation of cloud droplets and affecting cloud albedo. Aerosol direct and indirect effects are the largest source of uncertainty in humans' impact on the radiative balance of Earth (Farmer et al., 2021). The impacts of both direct and indirect aerosol effects are controlled by the concentration of aerosols in the atmosphere. The concentration of aerosols is based on their emission from the surface and eventual deposition back on the surface. Aerosols are emitted into the atmosphere in many natural ways: volcanoes, sea spray, wind, wildfires, plants, and animals. A variety of human activities also emit aerosols: industry, shipping, transportation, and agriculture. To understand the impact humans have had on our climate it is important to understand the processes which remove aerosols from the atmosphere: wet and dry deposition. A timeline of advances in our understanding of aerosols, instrumentation, and parameterizations will be discussed, as well as the impacts key papers have had on the field (**Figure 1**).

Dry deposition is an important process which removes particles from the atmosphere. Dry deposition refers to aerosols being directly taken up by a surface, while wet deposition involves clouds or precipitation collecting aerosols before falling to the surface. Although wet deposition removes many particles from the atmosphere dry deposition is still an important process for removing aerosols, especially in places and during times with little precipitation. As aerosols in the atmosphere approach the surface there are many ways they can be deposited on a surface, such as impaction, interception, and Brownian diffusion. Dry deposition removal mechanisms, such as those listed, are influenced by the aerosol's properties, meteorological conditions, and properties of the surface. Real-world measurements can guide our parameterizations; parameterizations which describe how dry deposition occurs over various conditions.

Alongside flux measurements, many methods have been implemented to attempt to measure and gain insight on dry deposition (Hicks, 1986). This paper discusses several approaches to measuring deposition and how measurements and theory at the time compare. Surrogate surfaces, containers which collect particles as they settle, have been used to measure particles deposition. However, questions are raised about how well these surfaces emulate environmental surfaces and how to compare upwards movement of particles to downwards. The 1986 paper by Hicks also explores micrometeorological methods like concentration gradient and flux methods. Importantly, “flux-measurement methods yield data on the rate of turbulent exchange across a quasi-horizontal plane...” (Hicks, 1986). Flux measurements do not directly measure particles depositing on or emitting from a surface. Connecting the exchange of particles between the atmosphere and surface to measurements of exchange above the surface requires many measurements with consistent metrological conditions and appropriate sites to collect data over.

Developments in aerosol flux measurements have occurred, contributing to a greater understanding of aerosols in our atmosphere and their removal processes. In 1977 flux measurements were preformed over moderately vegetated surface (Wesely et al., 1977). It was demonstrated that flux measurements could be performed to find a deposition velocity. However, this study is limited by instrument capabilities, instruments had slow response times and lots of periods of data collection could not be used as noise was too great. Additionally, only particles between 50-100 nm were measured and were not differentiated. Other studies were also limited in their ability to differentiate particles based on size, which is important as deposition velocity is dependent on particle size (Ruijrok et al., 1995). Greater time and particle diameter resolution would allow improved measurements. Sophisticated instrumentation has allowed for this, 10 Hz measurements of particles between 60-1000 nm were performed over a forest and grassland surface (Emerson et al., 2020). This study also used the instruments' ability to differentiate particles based on size to further improve measurements. As methods to measure aerosols have improved and more data has been collected the models used to describe the behavior of dry deposition have been updated.

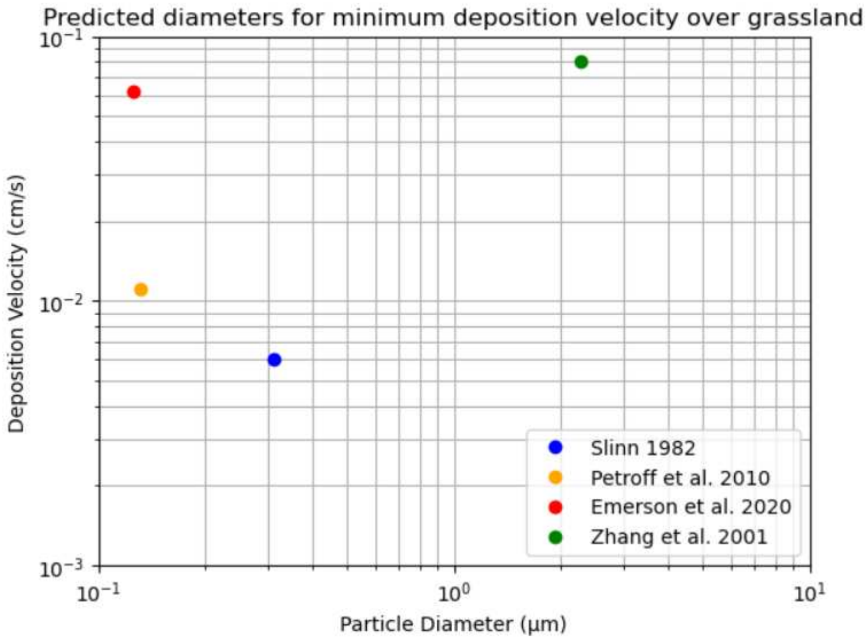

Figure 2. Different models predict the diameter of particles at which the minimum deposition velocity will occur. Minimums taken from predictions over grassland from the papers Slinn 1982, Petroff et al. 2010, and Emerson et al. 2020.

Models for aerosol dry deposition have been continuously iterated upon as conceptions of what drives deposition change and as new measurements are produced. A parameterization was developed to model dry deposition over vegetated surfaces (Slinn, 1982). The parameterization, amongst other factors, considers momentum of particles and the surface characteristics to predict deposition velocities of particles based on size. This model was a basis for future models developed for the same purpose, such as parameterizations by Zhang et al. in 2001. This model considers the variation in surface types across Earth by having 15 categories for land type and 5 for seasonal changes (Zhang et al., 2001). This allows the 2001 model to be more applicable across differing surface landscapes while the 1982 model is intended to be used only over vegetation. However, the 2001 model has a simplified term describing interception, especially when compared to later parameterizations. A more complex parameterization was created, based on the Zhang et al., 2001 model, it features a more complex term for interception and even greater specificity for land types with 26 land categories (Petroff et al., 2010). With greater specificity and more complex parameterizations it is more challenging to apply to existing aerosol models. Another paper, based on the 2001 parameterization by Zhang et al, created updated parameters from many new measurements, including their own (Emerson et al., 2020). The modifications to the Zhang parameters in the Emerson parameterization are in better agreement with recent measurements. As seen in F**igure 2**, the Petroff and Emerson parameterizations both account for a greater role of interception than the Zhang parameterization, which generally results in a shift of where the minimum deposition velocity occurs, from larger particles (~1.0µm) to smaller particles (~0.1µm).

The modern 2020 Emerson parameterization exists in its form due to prior parameterizations understandings of deposition and measurements which continuously improve. The impacts of aerosols and deposition on our climate can be more accurately understood and uncertainties can be reduced because of these modern models. However, improving measurements and creating new datasets over the variety of Earths landscapes will further our understanding of these processes and improve our climate predictions. For example, models from the Zhang parameterization appear to overestimate the impact of aerosols indirect and direct effects over the oceans (Emerson et al., 2020). Yet, more measurements over the ocean need to be performed to further refine our models. This is true for a variety of landscapes across Earth, so that models can be iterated based on empirical data.

CHAPTER 2 – OVERVIEW OF METHODS AND DATA PREPARATION

Introduction to Methods

Aerosol dry deposition velocities are calculated by first measuring fluxes of aerosols with the eddy covariance technique. Aerosol fluxes and concentrations are used to determine deposition velocities after the measurement takes place. The FALCON project seeks to make aerosol flux measurements over various types of surfaces, so far three surface types have been measured over: grassland, forest, and ocean. A sonic anemometer and a portable optical particle spectrometer (POPS) are operating at each site. Together these instruments make the measurements necessary for the eddy covariance technique. Raw anemometer and POPS data are processed and formatted so EddyPro can be used to calculate fluxes.

Measuring Aerosol Fluxes with The Eddy Covariance Technique

The eddy covariance (EC) technique has been used to improve scientists' understanding of surface-atmosphere exchange, especially for gases like carbon dioxide, methane, and water (Baldocchi, 2020). In addition to gases, EC can be used to find properties of surface-atmosphere exchange of aerosols, relevantly deposition velocity (Vdep). Turbulent airflow is composed of many eddies, which move packets of air, and aerosols, in three dimensions. For deposition and emission processes the vertical component of wind velocity (w) is considered. The quotient of vertical flux (Fc) and aerosol concentration (c) is the exchange velocity (Vex) between the surface and atmosphere.

$$V_{ex} = \frac{F_c}{c} \qquad \text{(Eq 1)}$$

A positive exchange velocity, and a positive flux, describes emission while a negative value is deposition. Often the vertical exchange of aerosols is described in terms of deposition velocity.

$$V_{dep} = -V_{ex} \tag{Eq 2}$$

Vertical flux can be equal to the covariance, or combined variability, of vertical wind speed and aerosol concentration, or some other scalar quantity of interest (Baldocchi, 1988).

$$F_c = \overline{w'c'} \tag{Eq 3}$$

Calculating fluxes requires vertical wind speeds and aerosol concentrations to be measured over an averaging period, 30 minutes is common, to capture the information from large eddies. Deviations from the average are also measured, requiring rapid measurements, faster than 5 hertz, to capture information from even very small eddies. In addition to instrumentation requirements, key assumptions about the surface must be accounted for by site selection and preparation. The surface being measured must be horizontally homogeneous and there must not be aerosol sources or sinks above the surface. The measurement must be made in the constant flux layer, the height of which depends on the height of the surface canopy. (Burba 2021 textbook).

Site Description

FALCON has collected eddy covariance data from 3 sites. Measurements over forest surfaces began on 15 May 2024 and are ongoing at Chestnut Ridge (CHS) in eastern Tennessee (35°57’31.4” N, 84°17’14.9” W). The altitude at CHS is ~413m and measurements are recorded at a height of 30m, below there is a canopy of broadleaf trees extending 15m. Ocean surface measurements took place from 17 April to December 19 of 2024 off the Ellen Browning Scripps

Memorial Pier (SCR) in San Diego (32°52’01.9” N, 117°15’26.8” W). The surface of SCR is at sea level and measurements are recorded ~13m above the surface, measurements are recorded on the westernmost point of the pier. The pier extends 335m east, at ~300m the shore is reached. Data collected over a grassland began on 12 December 2024 and is ongoing in northern Colorado’s Pawnee grasslands (PAW) at the semi-arid grasslands research center (40°48’30.9” N, 104°46’37.1” W). The altitude at PAW is ~1650m and measurements are made 1.8m high because the canopy height is relatively short. The canopy was measured to be 0.1m at one point but will change seasonally. The surfaces for each site are shown in **Figure 3**.

Figure 3. Surface types at FALCON measurement sites.

Each site has a sonic anemometer and POPS to make measurements. The sonic anemometer points in the direction of predominant wind and the POPS sits behind the sonic anemometer in a way that minimizes vertical displacement between the two instruments. Displacement between the sonic anemometer sensor and the POPS inlet displacement varies by site.

Instrument Description and Calibration

Assuming a site satisfies the requirements for EC, a sonic anemometer and POPS are used to measure wind velocity and aerosol concentrations, respectively. The POPS categorizes aerosols by size in a range of 120-3000nm into one of 9 bins. The edges of each bin vary at each measurement site but generally increase in width as the diameter increases; bin 1 might be 120-140nm and bin 9 might be 2400-3000nm. Grouping the aerosol concentration by size means the fluxes and Vdep are also calculated for each size range. For aerosols with diameters between 140-3000nm POPS ability to measure concentration and differentiate size is comparable to SMPS (Gao, 2016). Aerosol concentration and aerosol size measurements are both considered with the POPS calibrations.

Handix Scientific performs three calibrations on the Portable Optical Particle Spectrometers (POPS) before we receive them. A flow calibration ensures the pump pulls as much air as the POPS onboard computer expects, which is important for turning aerosol counts into concentrations. A calibration with a differential mobility analyzer compares the size of aerosol filtered for with the size of aerosols the POPS measures. Finally, a calibration with 5 different-sized PSL nanospheres confirms the POPS will correctly determine the size of the PSLs.

Before POPS are deployed in the field they are calibrated once more. Details on performing and processing this calibration are included in the POPS Calibration Guide. The final product of the calibration is a Mie curve, unique to each POPS at each field site. The Mie curve correlates scattering amplitude to aerosol diameter. This allows aerosol sizes to be determined and aerosols to be “binned” into 1 of 9 size bins. Fluxes will be calculated for each size bin, meaning Vdep will also be found for each size range. EddyPro 7, developed by LI-COR Biosciences, is software that processes each bin into vertical fluxes.

Pre EddyPro Data Processing

The field set-up to measure fluxes of aerosols is described in the FALCON Setup Guide (Peña, 2023). Preparing hardware, mounting instruments, configuring the on-site computer, retrieving data, and more are explained in that document. Preparing data to be processed by EddyPro requires two scripts, POPS_binary_extract.py and flux_prepping.ipynb. They are both described in the FALCON Setup Guide. Changes to both scripts have been made, so this document will also outline their use, including the updates made.

POPS_binary_extract.py converts binary files the POPS produces into CSV files while also applying the mie curve generated by the calibration, sorting each aerosol measurement into a size bin. This code should first be callable from anywhere on the computer, instructions for achieving this are in the FALCON Setup Guide. Using the terminal "POPS_binary_extract.py --help" will show options for using the code, that will also be explained below. Instructions 1-4 are also explained in the FALCON Setup Guide (Peña, 2023).

1. "-directory" path to the binary files downloaded from POPS
2. "--nbins" indicates the number of bins to sort aerosols into. The default is 16 bins. FALCON uses 9 bins
3. "-mie" path to the mie conversion table given in the python_scripts folder. Or generated following the POPS calibration
4. "--multiproc" doesn't need an argument. If typed, it will process the binary files in batches of four using four processors

Additionally, two more arguments must be made

5. "--logmin" is a minimum value of current the POPS will record when measuring aerosols. The value can be found in the POPS data directory, any CSV file that begins with "HK_" can be opened to find logmin
6. "--logmax" is the maximum value of current the POPS will record. Its value can be found in the same place as logmin

Running POPS_binary_extract.py in terminal example:

```
POPS_binary_extract.py --directory /folder/with/binary/data --nbins 9 --mie /file/containing/mie/curve --multiproc --logmin 1.75 --logmax 4.81
```

Flux_prepping.ipynb has been consolidated with the final step in preparing fluxes for EddyPro, eliminating the need to adjust the timestamp column to a format EddyPro recognizes each time. The FALCON Setup Guide (Peña, 2023) outlines what operations are performed (and even more information can be found on RickPena10's github). This document will only explain the steps to use this code. This code is intended to run on a Jupyter Notebook page and edits must be made to the second and last kernel. Run the code one kernel at a time, allowing loading bars to complete before continuing to the next kernel. When flux_prepping.ipynb has completed successfully there should be a folder named "EP_Ready" containing .txt files ready to be processed with EddyPro.

Second kernel edits:

1. "data_dir" path to the folder containing raw anemometer data and processed 10 Hz POPS data. Usually the flux_prep folder
2. "time_zone" time zone of the computer that is logging anemometer data. Not necessarily the time zone at the site. Usually UTC

3. “anemometer_type” type of anemometer used.
4. “loc” location code assigned to each site. SCR for Scripps Pier, CHS for Chestnut Ridge, PAW for Pawnee Grasslands

Last kernel edits

7. “folder_path” path to anemometer+POPS synced files with concentration data. Should be the same path in “data_dir” but including ./Anem_POPS_conc_csvs/ at the end

Overview of Data Processing

Processing raw POPS and anemometer files to a format that EddyPro can read and calculate fluxes from requires several steps. Calibrating a POPS before it makes field measurements is the first step. After raw data is collected two Python scripts are used to prepare data for EddyPro. First calibrations are applied to sort aerosols into bins while sampling the data down to 10 Hz. Then POPS and anemometer files are time-synced and combined into perfect 30 min 10 Hz files. Ensuring files are processed and formatted consistently at each step minimizes errors in scripts, unexplained gaps in data, and confusion about next steps in processing. Consistent methods of processing data are especially important when collecting large datasets where it can be easy to lose track of data. **Figure 4** shows the steps FALCON uses to process data, describing the file contents at each step and naming the scripts and programs used to process data.

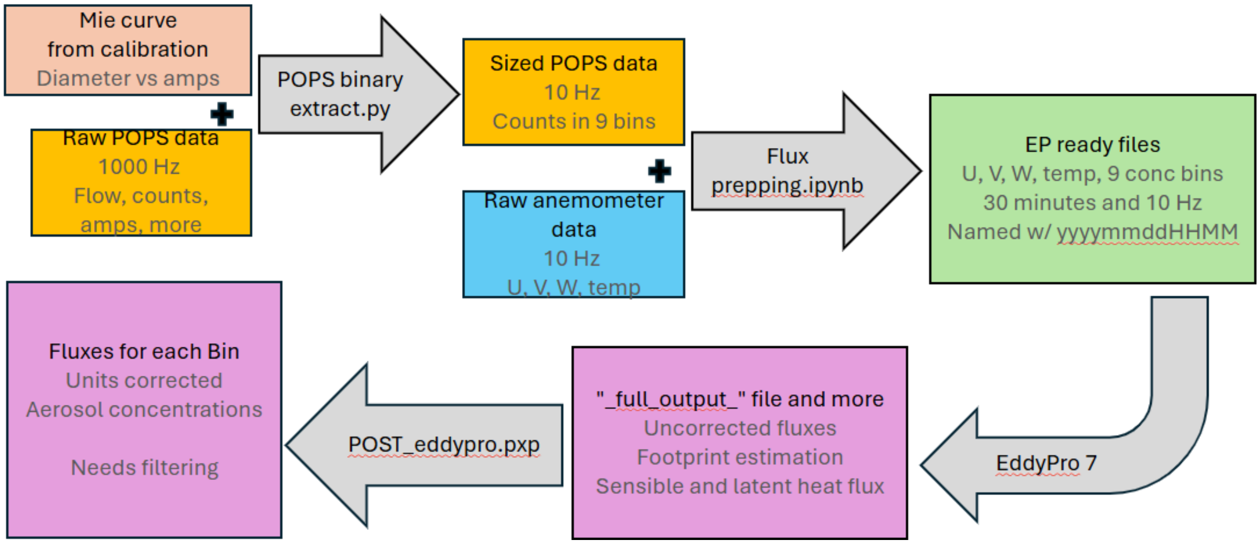

Figure 4. FALCON data processing flow chart.

CHAPTER 3 – PRELIMINARY ANALYSIS AND RESULTS

Introduction to Analysis

Fluxes of aerosols are calculated with EddyPro before being corrected with a custom Igor Pro script. Time series of fluxes and average aerosol concentrations are used to calculate deposition velocities after any filters have been applied. Example data collected at Pawnee grasslands is shown and deposition velocities for bins are calculated. Deposition velocities are filtered to exclude instances of rain or snow as well as low turbulence. Frictional velocity or u* describes how turbulent the flow of air is, excluding times of low u* increases confidence in flux measurements as eddy covariance assumes turbulent flow is what transports aerosols vertically at the point of measurement. EddyPro produces frictional velocity calculations from U, V, and W wind speeds while processing fluxes.

Processing with EddyPro

EddyPro is open-source and free software that processes eddy covariance data to produce gas fluxes. The EddyPro FALCON document outlines how to use EddyPro to process aerosol fluxes. The output of EddyPro is several folders and files, the most important file moving forward is named “eddy_pro_TITLE_OF_PROJECT_full_output_DATE.csv” where the parts in all caps are unique. Open this CSV in Excel to see any errors (-9999 values) readily. It's recommended to check that there are no error values for any of the times with data in the “EP_Ready” folder. If there are errors at inappropriate times, re-run EddyPro and ensure the

Basic Settings tab is filled out correctly. Once flux data is confirmed to match the dates/times from wind and particle data the next step is to process with POST_eddypro.pxp.

Post EddyPro Data Processing and Filtering

POST_eddypro.pxp is an Igor Pro experiment (run with Igor 8 not Igor 9) that converts fluxes from each EddyPro output into the units $\#\bullet cm^{-2}\bullet s^{-1}$ which is appropriate for aerosol fluxes because concentration is calculated as a number concentration ($\#\bullet cm^{-3}$) and V_{dep} is often represented in $cm\bullet s^{-1}$. Additionally, bins assigned to H_2O flux on EddyPro need to be multiplied by 1000 because of the units EddyPro assigns water vapor. POST_eddypro.pxp also organizes and renames many of the indices produced by EddyPro so they represent our data. To use this script:

1) Launch POST_eddypro.pxp with Igor Pro 8
2) Run "ep_extract_all()" in the command window
    a. Navigate to and select the "full_output" file for bins 123
    b. Change the date and time column to text format before submitting
3) Repeat step two with the "full_output" files for bins 456 and 789T, "ep_extract_all()" will automatically open file explorer thrice to select three files
    a. If files are selected in the wrong order close Igor Pro without saving and try again
4) Run "time_unit_corr()" in the command prompt
5) Mean concentrations and corrected fluxes are waves in "Flux_waves" under "EP_results"
    a. Deposition velocities are the quotient of each 30-minute flux value and mean concentration
    b. Downwards fluxes and deposition velocities are positive (negative results are upwards fluxes)

With deposition velocities calculated for each 30-minute period (or row in the dataset), various filters should and could be applied to the data. Circumstances which invalidate the eddy covariance method should be tracked so those inappropriate periods are filtered. Filtering times during active precipitation or when instruments are moved (like when the boom was reeled into the pier at SCRIPPS) is necessary. Data could also be filtered to exclude times of low turbulence, u* is a measure of turbulence and is included in EddyPro's output. Filtering by wind direction is also possible; to exclude wind from the back 20° of csat3b sonic anemometers data as it is less reliable or exclude certain regions of the flux station's fetch (area on the surface that can be measured). Filtering will look different depending on the circumstances of the site. Whole days, or longer, may be filtered or only a single 30-minute period. Ensure the times being filtered are accurate considering FALCON-produced fluxes should always be in UTC but data used for filtering may be in local time and may include time switches from daylight savings.

Pawnee Grassland Aerosol Fluxes and Concentrations

About one month of eddy covariance data has been collected at the Pawnee grasslands site during winter. Time series of aerosol fluxes for bins 2-8 are shown in **Figures 5, 6** concentrations over the same time and bins are shown in **Figures 7, 8**. The flux and concentration values below are used in calculating the deposition velocities for their respective bins.

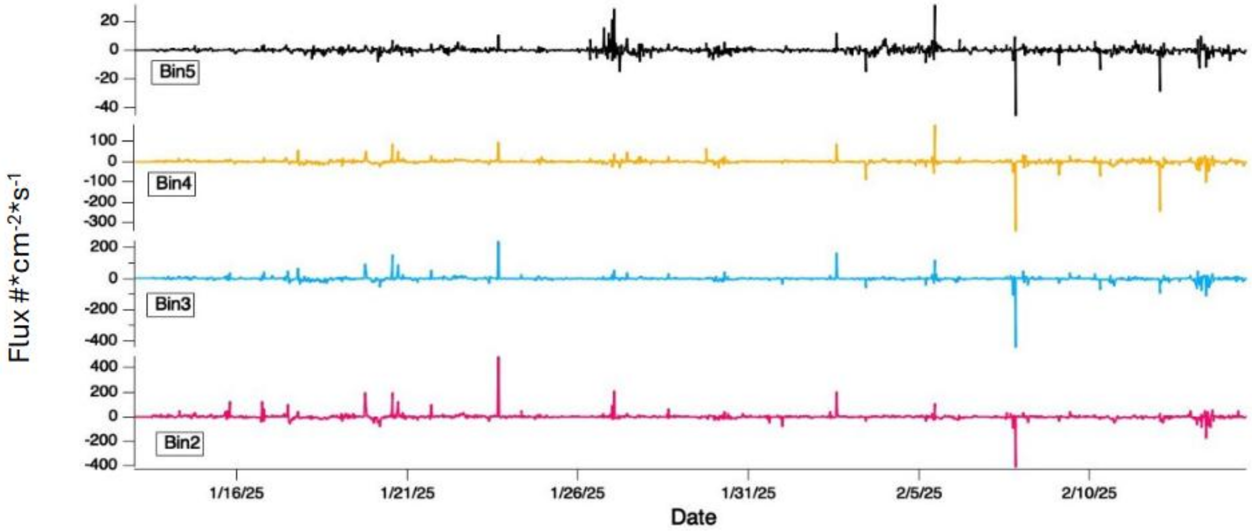

Figure 5. Pawnee grassland aerosol fluxes of smaller size bins from Jan 13 – Feb 14, 2025.

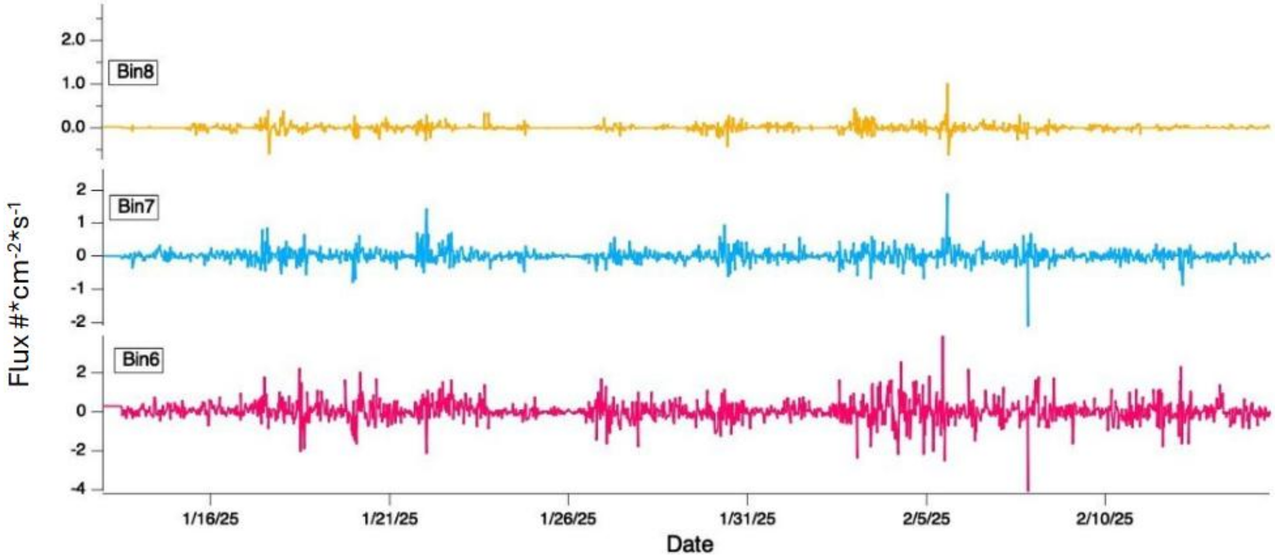

Figure 6. Pawnee grassland aerosol fluxes of larger size bins from Jan 13 – Feb 14, 2025.

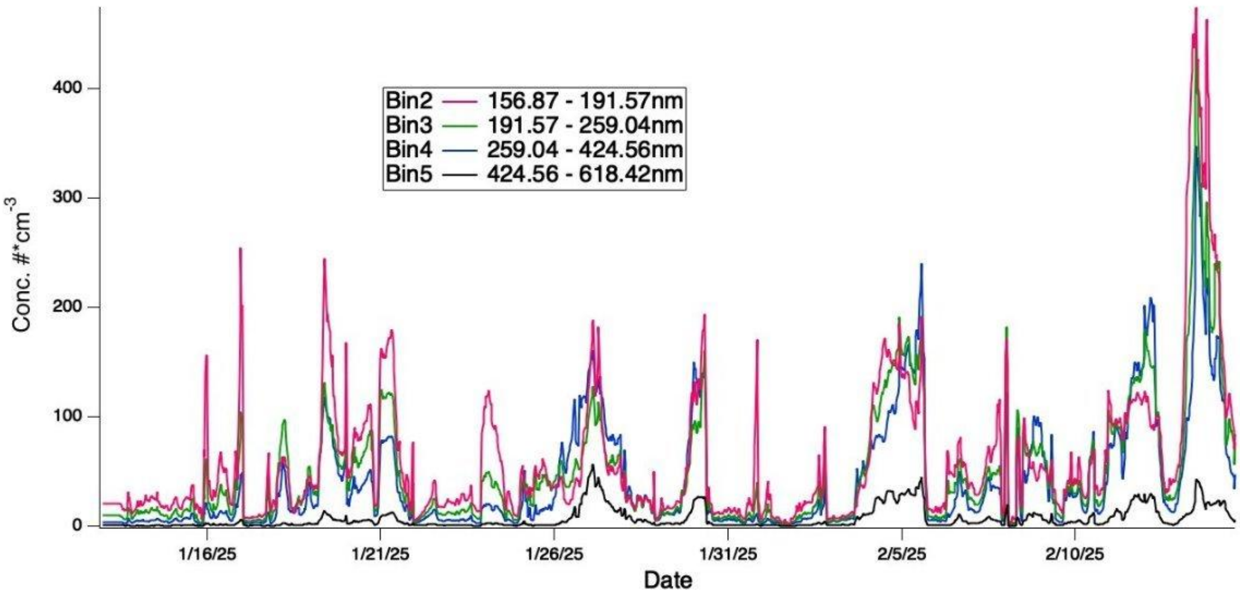

Figure 7. Pawnee grassland aerosol concentrations of smaller size bins from Jan 13 – Feb 14, 2025.

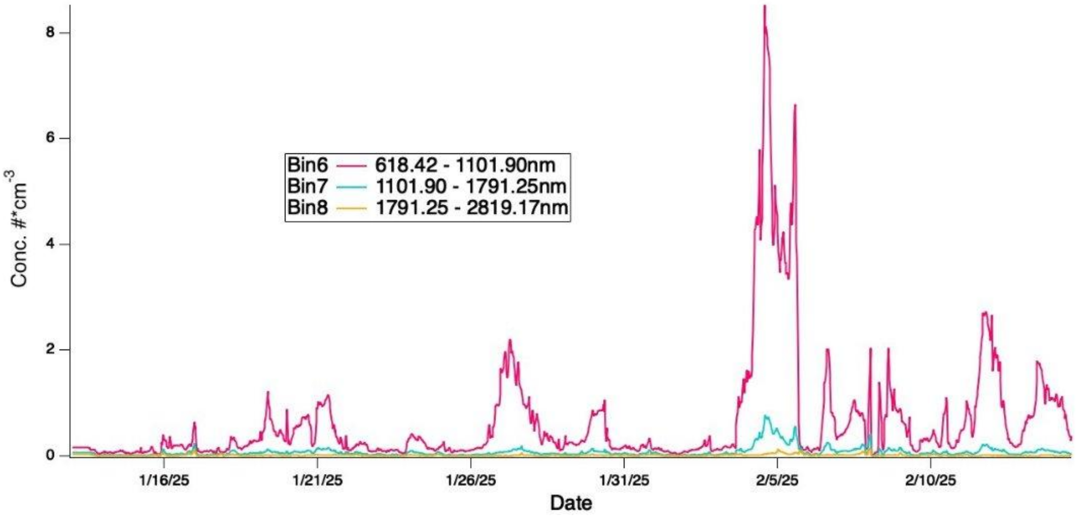

Figure 8. Pawnee grassland aerosol concentrations of larger size bins from Jan 13 – Feb 14, 2025.

Early Pawnee Grassland Deposition Velocity Results

Aerosol concentrations and fluxes at Pawnee grassland are used to calculate deposition velocities for each size bin. The results of which are shown in **Table 1**. Positive values are deposition velocities (downwards direction), and negative values imply an upwards movement of aerosols.

Table 1. Pawnee Grassland size dependent aerosol deposition velocities. Collected from December 2024 to February 2025 and filtered to exclude precipitation events.

Bin and Aerosol Size Range (nm)	Average V_{dep} (cm•s^{-1})
Bin 1: 132.44 - 156.87	-0.16
Bin 2: 156.87 - 191.57	-2.3×10^{-2}
Bin 3: 191.57 - 259.04	-3.0×10^{-2}
Bin 4: 259.04 - 424.56	4.1×10^{-2}
Bin 5: 424.56 - 618.42	4.3×10^{-2}
Bin 6: 618.42 - 1101.90	0.45
Bin 7: 1101.90 - 1791.25	0.10
Bin 8: 1791.25 - 2819.17	0.27
Bin 9: 2819.17 - 3959.50	-0.62
Total Bins 132.44 - 3959.50	-0.12

The values in **Table 1** are preliminary and need more analysis and filtering. However, by following this guide and with the appropriate calibration curve and raw Pawnee data (from Dec 2024 to Feb 2025) **Table 1** can be reproduced. **Table 2** shows the deposition velocities for some bins after filtering for high turbulence and removing outliers that are beyond 1.5 times the interquartile range from the 25th and 75th percentile. Overall, about 80% of the original dataset is removed with most remaining measurements from the daytime.

Table 2. Pawnee Grassland deposition velocities measured from December 2024 to February 2025. Filtered for no precipitation, high turbulence ($u^* \geq 40$ cm/s), and excluding outliers.

Bin #	Average V_{dep} (cm•s^{-1})	Standard deviation (cm•s^{-1})
Bin 1: 132.44 - 156.87	-0.10	0.47
Bin 2: 156.87 - 191.57	-6.2×10^{-2}	0.44
Bin 3: 191.57 - 259.04	1.5×10^{-3}	0.52
Bin 4: 259.04 - 424.56	3.0×10^{-2}	0.60
Bin 5: 424.56 - 618.42	0.13	1.1
Bin 6: 618.42 - 1101.90	0.60	3.4
Bin 7: 1101.90 - 1791.25	1.1	6.1
Bin 8: 1791.25 - 2819.17	0.57	2.4

Bin 9 is not shown in **Table 2** because the diameter range collected in this bin exceeds the POPS size detection in this case. Processed deposition velocities calculated from Pawnee data are compared to other grassland measurements and models in **Figure 9**. Bins 3-8 are plotted with each bin's midpoint as a marker.

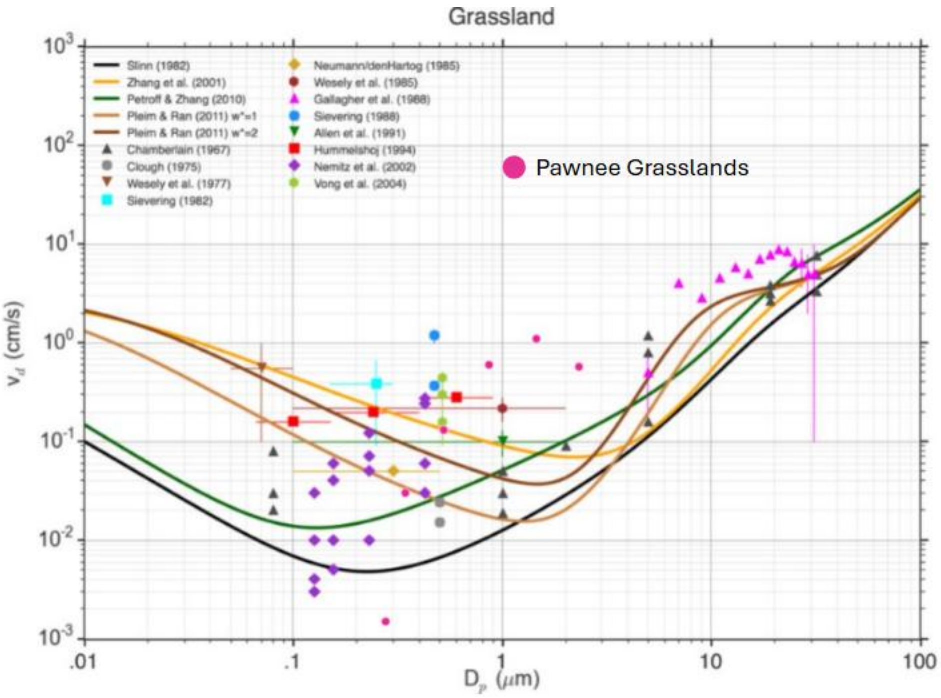

Figure 9. Winter Pawnee grassland deposition velocity measurements. Filtered for no precipitation, high turbulence (u* ≥ 40 cm/s), and excluding outliers. Comparision to other grassland measurements and models. Figure adapted from Saylor, 2019.

Overview of Preliminary Results

Aerosol fluxes and average concentrations at Pawnee grasslands are used to calculate deposition velocities for each size bin that FALCON sorts aerosols into. Deposition velocities are represented as a time series in Igor, where the above data processing procedure ends. Filtering eddy covariance data is often necessary for valid measurements but also can help identify what factors influence deposition velocity. In what ways do atmospheric turbulence, temperature, time of day, seasons, etc influence deposition velocities? This question can be explored with data collected at each site because all POPS and anemometer data are time synced and saved in Igor.

CHAPTER 4 – CONCLUSIONS AND FUTURE WORK

In conclusion, EddyPro and custom scripts have been used to calculate vertical aerosol fluxes and deposition velocities from aerosol counts collected by a POPS and wind velocities measured by sonic anemometers. POPS are calibrated to generate mie curves that relate the signal produced by POPS to aerosol size. Then two scripts are used to prepare raw files for EddyPro. Mie curves are applied to POPS measurements after data collection and the measured aerosols are sorted into one of nine size bins. Aerosol concentrations are time-synced with wind data at 10 Hz and placed into exactly 30-minute files while naming the files according to EddyPro's date/time convention. EddyPro processes aerosol data 3 bins at a time as if it is processing gas fluxes. One more script is used to rename columns and correct the units of EddyPro's output to be consistent with fluxes of aerosols. Deposition velocities are calculated from each 30-minute flux period and the average aerosol concentration from the same period. However, filtering is necessary to exclude deposition velocities calculated from periods where the eddy covariance technique is inapplicable or to increase confidence in data. Once filtered, deposition velocities for each size bin can be averaged.

I have documented a procedure for performing single-point POPS calibration. From experience doing calibrations and using their output, I have included troubleshooting steps. Minor changes to how calibration curves are applied to raw POPS data have also been made so curves match the POPS range of detection. Processing files from their raw state to a point where EddyPro can use them required three scripts and generated a lot of files that only got used once. By adding to the flux_preping.ipynb this part of processing has been shortened to two scripts and saves some storage by eliminating 1 file type that used to be produced. Many of the inputs for EddyPro were determined by me, including what time-lags to use and many of the options in Advanced Settings. Although many options in EddyPro do not influence the actual fluxes so more decisions about what to includ in the output of EddyPro can be made.

Calculating fluxes and preliminary deposition velocities is only the beginning of understanding what these measurements mean for aerosol dry deposition over their respective surfaces. More work to better understand the capabilities of the FALCON approach to measuring aerosol fluxes can be done. Uncertainty in aerosol diameters measured by POPS is not fully understood. The extent to which POPS measurements change after being deployed in the field for several months is also not known. Performing calibrations on POPS after field deployment is a start to answering the above concerns. Tests comparing the outcomes of various filters over the same dataset can inform additional changes to how FALCON data should be filtered. For example: does filtering out times of low turbulence or certain wind directions increase confidence in results. Analysis to understand the apparent upward fluxes of aerosols in some size ranges would also be informative. Producing size-resolved deposition velocities over various landscapes allows an investigation of what processes determined the value of deposition velocities measured.

FALCON measures fluxes of aerosols in ways that are unique from other studies that seek to determine aerosol dry deposition velocities. The FALCON project makes use of the POPS, a relatively low-cost instrument, to make aerosol measurements. This allows more measuring sites (with different surface types) to be active at once because more instruments can be purchased. Additionally, because FALCON sites are a network, a scientist does not have to always be close to the site to record data. This means several months of eddy covariance data can be collected at a site before the site needs to be serviced. FALCON can generate long datasets at several sites for less cost than other means of measuring deposition velocities.

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APPENDIX A – POPS CALIBRATION GUIDE

Goal of calibration

The goal of this POPS calibration is to generate a Mie curve unique to each POPS at each field site. Mie curves relate aerosol diameter to scattered light intensity, or something proportional to light intensity such as the current produced by a PMT. A Mie curve will be applied to POPS data after the fact to turn signal to diameter.

Preparing calibrant

PSLs from Thermo Fisher Scientific, and certified by NIST, have a known diameter and refractive index (RI). In a container with ~50 mL DI water add 3-5 drops of PSLs. The exact concentration of aerosols in the water is not crucial for the calibration, however, too few drops of PSL in water will lead to low aerosol signal and make it hard to find the scattering amplitude of interest. The container of DI water and PSLs can be placed in a sonicator to suspend the PSLs and minimize them sticking together. After ~5 min in an ultrasonic bath the mixture is ready.

Calibration set-up description

An atomizer or PAGS can be used to suspend the aerosols in the air. Once afloat, aerosols flow to a 3-way intersection: the direction of the PAGS/atomizer, a filter which stops any aerosols and releases clean air, and a direction which leads to a drying tube. The PAGS/atomizer pushes air to the POPS while the POPS is pulling air. Because the POPS pulls less air than the PAGS/atomizer pushes the excess air is filtered as it safely leaves the system via the filter. The aerosols which are not filtered make it to the drying tube (silica beads are used as the desiccant) this removes water on the PSLs. If this step were skipped, aerosols larger than the expected size would reach the POPS due to hydroscopic growth. Then the aerosols can then flow into the

POPS inlet to be measured. Optionally, after the drying tube and before the POPS it is possible to run the aerosols through a differential mobility analyzer (DMA) to further exclude any incorrectly sized aerosols. Using a DMA is especially recommended if the POPS is measuring extra peaks other than what is expected from the PSLs, peaks around 180nm are common and are likely due to surfactants. Whether a DMA is used or not, the POPS should be plugged in, powered on, have an SD card in, and be connected to a computer via ethernet (**Figure 3**).

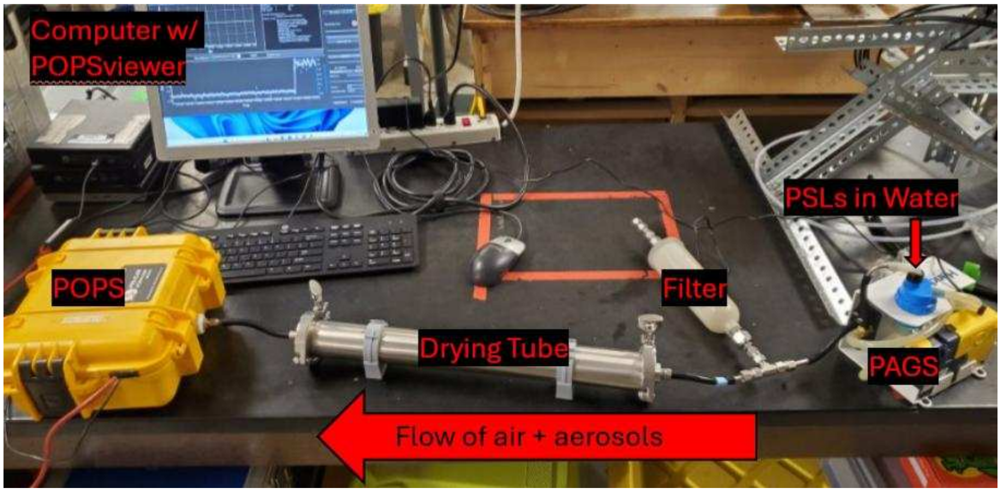


Figure 10. Single point POPS calibration setup.

Using POPSviewer to monitor calibration

POPS real time viewer is software made by Handix Scientific and can be downloaded here. Using this software, you can monitor the calibration live. A histogram of aerosol diameter vs counts is displayed in the center of the application, 16 bins are present in this histogram by default. Change this to at least 200 to see the aerosol size measurements more clearly. The RI on the software should also be changed to 1.615 as this is the RI of the calibrant PSLs. Both settings can be changed in the “Advanced” tab and selecting the right-hand column to edit.

laser feedback	620.22	🟥	620.22	🟩
laser power	949.89	🟥	949.89	🟩
auxiliary temp	25.59	🟥	25.59	🟩
input dc power	11.48	🟥	11.48	🟩
laser current	2.87	🟩	2.87	🟩
flow set	2.15	🟩	2.15	🟩
blstart	30000.0	🟩	30000.0	🟩
th_mult	3.0	🟩	3.0	🟩
number of bins	16.0	🟩	200	🟩
logmin	1.75	🟩	1.75	🟩
logmax	4.81	🟩	4.81	🟩
skip	0.0	🟩	0.0	🟩
minpts	8.0	🟩	8.0	🟩
maxpts	255.0	🟩	255.0	🟩

Figure 11. POPSviewer advanced tab settings.

Gain adjustment on POPS

The main peak visible on POPS real time viewer should be from your calibrant PSLs. Use a small flathead screwdriver to manually adjust the gain on the POPS to center this peak around your known diameter (usually 500nm or 510 nm). The screw to turn is above the PMT, to the left of a serial cable, and on a tiny blue square. There is a delay between the adjustment and the output on the software so slowly making small adjustments is recommended.

Figure 12. Gain adjustment on POPS.

Collecting calibration data

Once you are confident with your gain adjustment allow 3-5 minutes of aerosol flow into the POPS, making note of this time as you will need it later. Then remove the flow of aerosols from the POPS and turn off the PAGS/atomizer to stop generating aerosols. Connect the POPS inlet to a filter, you should see counts go close to 0 within a few seconds. If it has been a minute and you see counts >10 then you likely have a leak. You can test the POPS with a filter before the calibration as well, but I always do it after the calibration to confirm.

When you are ready to process the calibration and generate a mie curve access the storage of the POPS onboard computer (you can ssh into the POPS or remove the SD card from the POPS and open it on your computer). Either way look for the folder that corresponds to the day you performed the calibration. It will be in media>uSD>Data and the folder is named with the convention Fyyyymmdd. F20250210 would be the name of the folder generated on February 10th, 2025. This is the folder that will be referenced throughout the rest of this document.

Processing calibration data

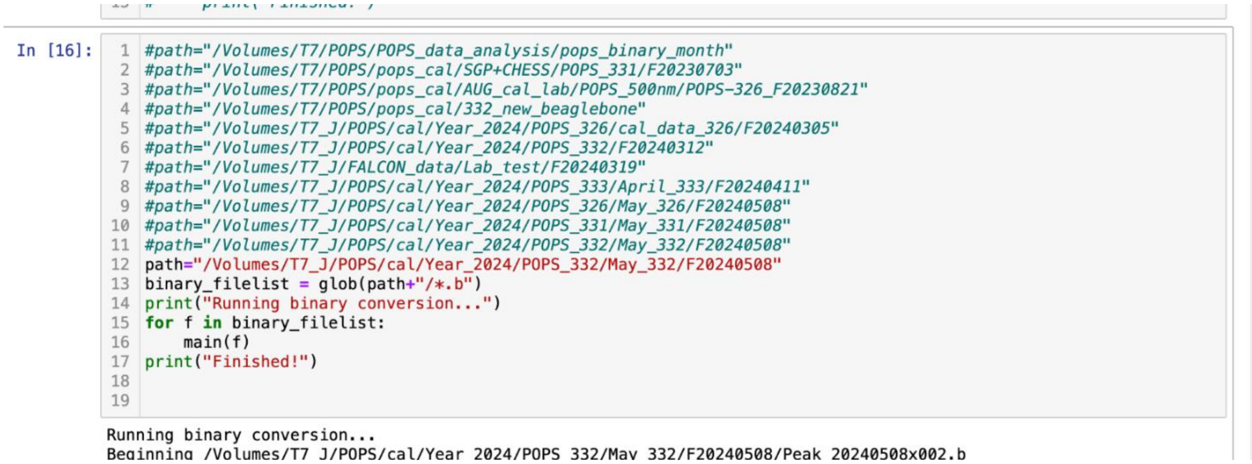
To create a Mie curve from the files generated by the POPS three pieces of code will be used. The computer that is doing this processing will require IgorPro 8 and python. Python code is intended to run in Jupyter Notebook.

All code mentioned in this SOP can be found in the Farmer group RStor:

farmerlab>Lab Resources>Software>FALCON>POPS_Calibration

Instructions for setting up the code on your computer are available as READMEs on RStor. Windows with Anaconda installed is recommended.

1) First turn binary files to CSV with “POPS_Binary_to_CSV-loop.ipynb” in jupyter notebook



```
In [16]:
#path="/Volumes/T7/POPS/POPS_data_analysis/pops_binary_month"
#path="/Volumes/T7/POPS/pops_cal/SGP+CHESS/POPS_331/F20230703"
#path="/Volumes/T7/POPS/pops_cal/AUG_cal_lab/POPS_500nm/POPS-326_F20230821"
#path="/Volumes/T7/POPS/pops_cal/332_new_beaglebone"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_326/cal_data_326/F20240305"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_332/F20240312"
#path="/Volumes/T7_J/FALCON_data/Lab_test/F20240319"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_333/April_333/F20240411"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_326/May_326/F20240508"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_331/May_331/F20240508"
#path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_332/May_332/F20240508"
path="/Volumes/T7_J/POPS/cal/Year_2024/POPS_332/May_332/F20240508"
binary_filelist = glob(path+"/*.b")
print("Running binary conversion...")
for f in binary_filelist:
    main(f)
print("Finished!")


Running binary conversion...
Beginning /Volumes/T7 J/POPS/cal/Year 2024/POPS 332/May 332/F20240508/Peak 20240508x002.b
```

Figure 13. POPS calibration file path to create CSVs.

a. Provide the path to the POPS folder you saved to your computer in the last kernel of this Jupyter Notebook (Pictured above)

b. Csvs will be made in same folder, named after the binary files: “Peak...x001.b” will generate “Peak...x001.csv”

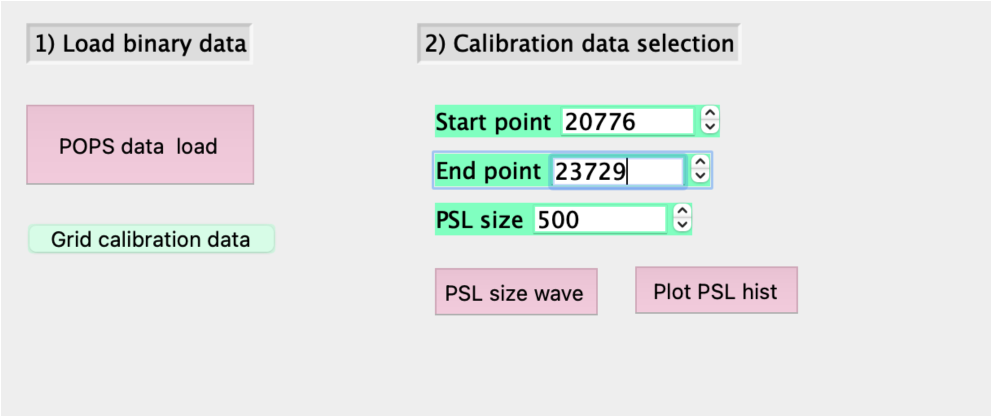
c. Once csv files are made you can move onto the next step

2) Find the peak scattering amplitude measured by POPS during calibration

a. Open “POPS_cal_toolkit.pxp” in IgorPro 8

b. Select “POPS data load” button and select csvs made from previous step

c. Select grid calibration data, if it worked new waves would be created



Start point 20776
End point 23729
PSL size 500

Figure 14. Loading POPS signals to curve fitting tool.

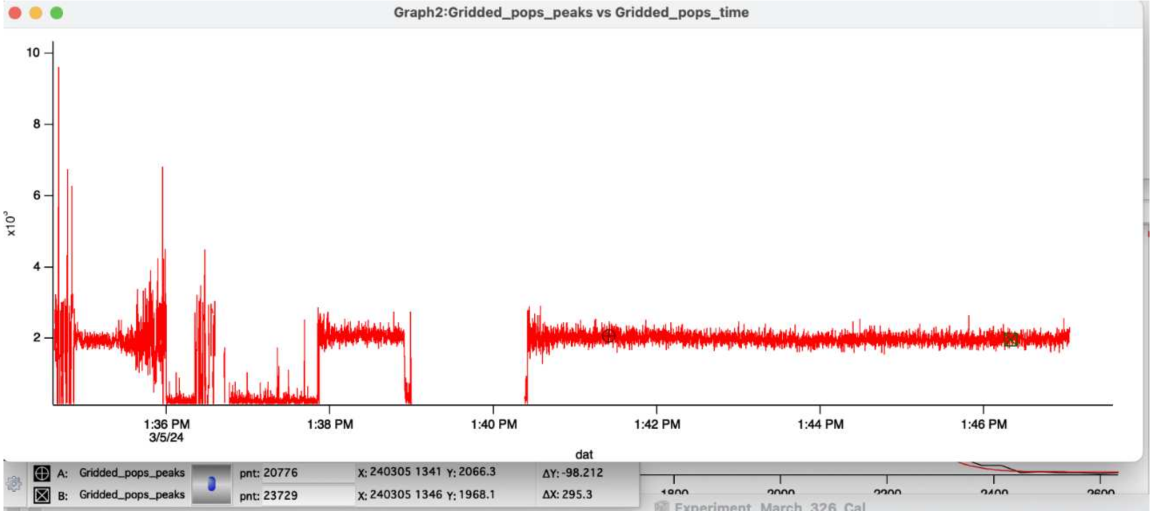
d. Ignore “matrix time” error if it shows

e. Create new graph in Igor

        i. Plot “gridded_pops_time” (x) vs “gridded_pops_peaks” (y)

f. Zoom in on time calibration was done, it may be obvious (flat and stable periods)

        i. If the calibration period is not obvious the x axis can be converted to HH:MM in Igor's plot settings to make it easier to tell



A: Gridded_pops_peaks pnt: 20776 X: 240305 1341 Y: 2066.3 ΔY: -98.212

B: Gridded_pops_peaks pnt: 23729 X: 240305 1346 Y: 1968.1 ΔX: 295.3

Figure 15. POPS calibration time series.

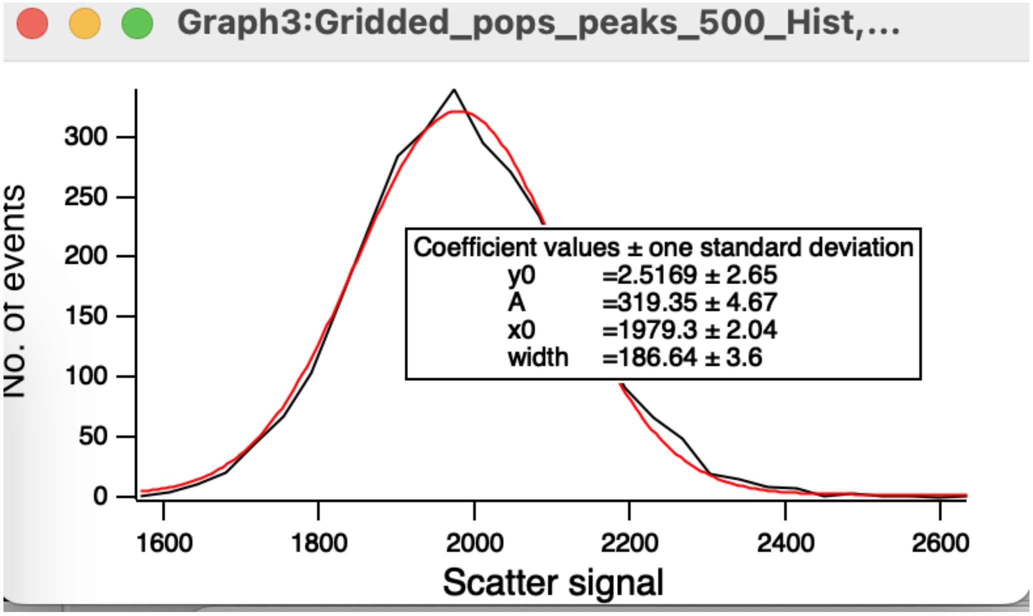
g. Click on plot, select ctrl+I

        i. Marker button A marks start of calibration, button B is for end, click and drag onto graph

h. The start and end points will be given in the pop up, put the start and end point back into the button window along with the PSL size

i. Select “PSL size wave” then select “Plot PSL hist”

        i. A calibration curve will be made



Coefficient values ± one standard deviation
y0 =2.5169 ± 2.65
A =319.35 ± 4.67
x0 =1979.3 ± 2.04
width =186.64 ± 3.6

Figure 16. Peak scattering amplitude for POPS calibration.

j. "x0" on this plot is peak scattering amplitude and should be noted for the next step

3) Generate a Mie curve from the values you now know

  a. Open Jupyter notebook "POPS_calibration_mie_curve_v2.ipynb"

  b. Several lines of this notebook need to be modified, they are listed below but the comments in the code should also tell you

  c. Some, but not all, variables will need to be changed:

    i. Dr: this is the diameter range the mie table will produce, keep as 100 to 5000

    ii. Ior: this is the refractive index, change to best match the aerosols of interest (1.588 may be a "reasonable" value in one site but not in another)

    iii. Single_pnt_cali_int: this is what x0 from igor should be

    iv. Single_pnt_cali_d: this is calibrated aerosol diameter (500 or 510 is normal for our calibrant)

        v. Single_point_cali_ior: this is the refractive index for the calibrant used (1.615 is for the PSLs)

   d. Run all lines from start until “df.to_csv(‘name_of_your_file’) and change the name to something normal
        i. I keep the RI, x0, and POPS number as part of my naming convention to keep track
   e. This will produce a mie table in the same directory that the code is running in
        i. If you want to apply smoothing to this mie table (you do) run the next kernel after making changes to the names of the files

The Mie curve you have generated will be applied to POPS data after collection. Instructions on applying the Mie curve to your data can be found in FALCON: A New Approach to Porcessing Fluxes of Aerosols section Pre-EddyPro Data Processing.

APPENDIX B – USING EDDYPRO FOR FALCON GUIDE

EddyPro (EP) is free and open-source software. This document is designed specifically for version 7.0.9. It is used for processing eddy covariance data, providing the user with fluxes. Although this software was not intended to process aerosol fluxes it can still be done! This document contains instructions on how aerosol fluxes are processed with EP. Most settings in EP have links which explain what the setting does so further experimentation on what the most useful settings are is possible.

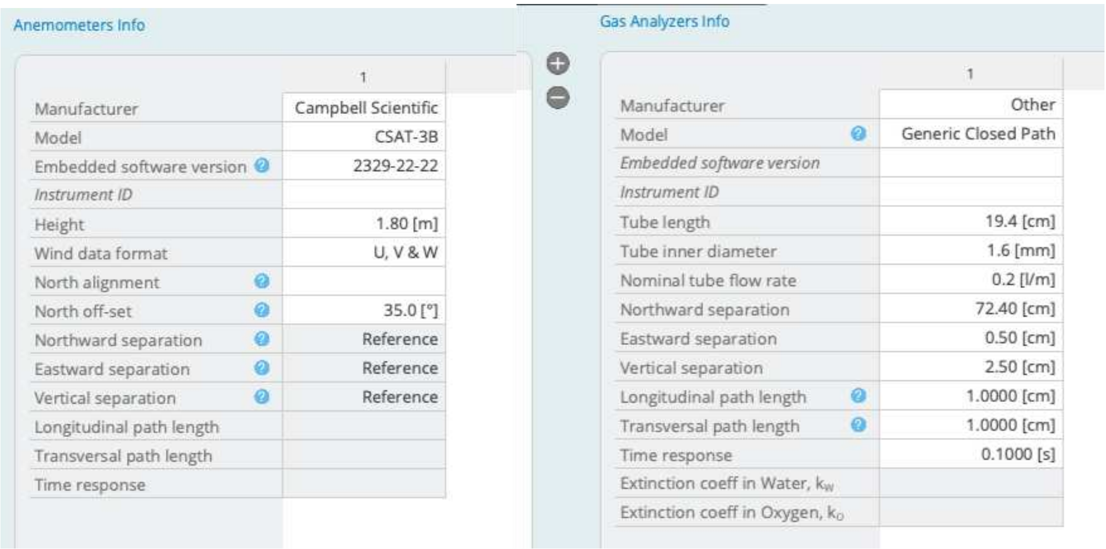
Project Creation

EddyPro processes up to 4 gas fluxes per run. When uploading data to EP aerosol concentrations are processed as if they were a gas. Aerosols are sorted into 9 bins before processing, so EP must be run three times to process each bin. In the first run bins 1, 2, and 3 are processed, then 4, 5, and 6, and finally bins 7, 8, 9, and total concentration are processed. EP is told bins 1, 4, 7 are CO_2 bins 2, 5, 8 are H_2O bins 3, 6, 9 are CH_4 and the total flux is N_2O. After processing the fluxes require a unit conversion to be appropriate for aerosol concentration. Project creation settings can be almost entirely automatically applied by creating and choosing a metadata file. Critical information for calculating fluxes is stored in the metadata files, including site and instrument specifications.

1) First, name your project. Naming your project according to the site and bins being processed is a good idea, like “PAW_bins123” for Pawnee
2) Select ASCII plain text as your raw file format
3) If you are using a metadata file load that in now

   a. By selecting load and navigating to where your metadata files are saved

4) If you loaded in a metadata file you should not need to make edits in step 5 and 6, but checking that settings were applied correctly is recommended
5) Under the station tab input, the correct site specifications (including canopy height, altitude, coordinates, and more)
6) In the instruments tab add information about both anemometer and POPS as below:
   a. The north off-set and height of anemometer change site by site
   b. The POPS info is under the gas analyzers info section because we are telling EP that we are calculating gas fluxes. Most POPS info is based on measurements of POPS inlet tube and will not need to be changed. Except for northward, eastward, and vertical separation which describe the distance between the POPS inlet and the center of the anemometer's measurement, this must be measured at each site



Anemometers Info

	1
Manufacturer	Campbell Scientific
Model	CSAT-3B
Embedded software version	2329-22-22
Instrument ID	
Height	1.80 [m]
Wind data format	U, V & W
North alignment	
North off-set	35.0 [°]
Northward separation	Reference
Eastward separation	Reference
Vertical separation	Reference
Longitudinal path length	
Transversal path length	
Time response	

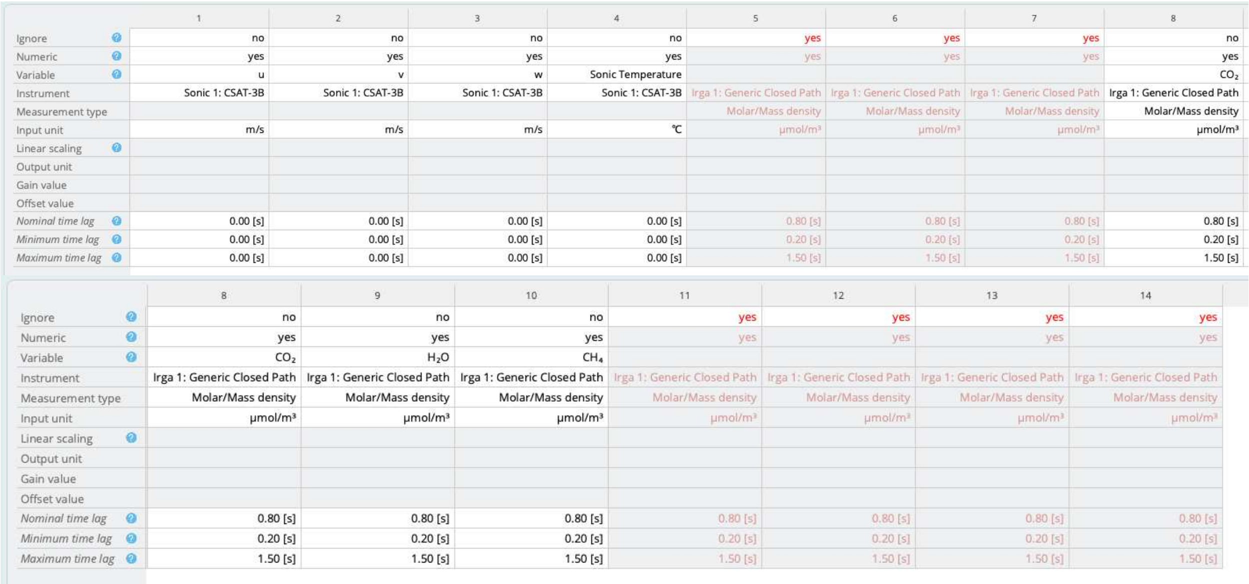
Gas Analyzers Info

	1
Manufacturer	Other
Model	Generic Closed Path
Embedded software version	
Instrument ID	
Tube length	19.4 [cm]
Tube inner diameter	1.6 [mm]
Nominal tube flow rate	0.2 [l/m]
Northward separation	72.40 [cm]
Eastward separation	0.50 [cm]
Vertical separation	2.50 [cm]
Longitudinal path length	1.0000 [cm]
Transversal path length	1.0000 [cm]
Time response	0.1000 [s]
Extinction coeff in Water, k_W	
Extinction coeff in Oxygen, k_O	

Figure 17. EddyPro instrument settings.

7) The raw file description tab will need to be edited for each run of EP (running bins 123, 456, and 789Total). This section tells EP how to read the .txt files we will provide it

    a. There are 14 columns total. Columns1-4 are anemometer data and 5-14 are POPS data. Keep anemometer columns the same for all runs and only change rows 5-14
    b. The screenshot below shows how the file description should look like when bins 4, 5, and 6 are being run. Setting the first row to ignore (yes or no) allows us to control which columns EP will process. Importantly, bins which are not being ignored need a variable (we use CO2, H2O, and CH4, and N2O)
    c. Finally, ensure the other rows match what is show below, i.e. measurement type, units, time lags, etc



	1	2	3	4	5	6	7	8
Ignore	no	no	no	no	yes	yes	yes	no
Numeric	yes	yes	yes	yes	yes	yes	yes	yes
Variable	u	v	w	Sonic Temperature				CO_2
Instrument	Sonic 1: CSAT-3B	Sonic 1: CSAT-3B	Sonic 1: CSAT-3B	Sonic 1: CSAT-3B	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path
Measurement type					Molar/Mass density	Molar/Mass density	Molar/Mass density	Molar/Mass density
Input unit	m/s	m/s	m/s	°C	µmol/m³	µmol/m³	µmol/m³	µmol/m³
Linear scaling								
Output unit								
Gain value								
Offset value								
Nominal time lag	0.00 [s]	0.00 [s]	0.00 [s]	0.00 [s]	0.80 [s]	0.80 [s]	0.80 [s]	0.80 [s]
Minimum time lag	0.00 [s]	0.00 [s]	0.00 [s]	0.00 [s]	0.20 [s]	0.20 [s]	0.20 [s]	0.20 [s]
Maximum time lag	0.00 [s]	0.00 [s]	0.00 [s]	0.00 [s]	1.50 [s]	1.50 [s]	1.50 [s]	1.50 [s]

	8	9	10	11	12	13	14
Ignore	no	no	no	yes	yes	yes	yes
Numeric	yes	yes	yes	yes	yes	yes	yes
Variable	CO_2	H_2O	CH_4				
Instrument	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path	Irga 1: Generic Closed Path
Measurement type	Molar/Mass density	Molar/Mass density	Molar/Mass density	Molar/Mass density	Molar/Mass density	Molar/Mass density	Molar/Mass density
Input unit	µmol/m³	µmol/m³	µmol/m³	µmol/m³	µmol/m³	µmol/m³	µmol/m³
Linear scaling							
Output unit							
Gain value							
Offset value							
Nominal time lag	0.80 [s]	0.80 [s]	0.80 [s]	0.80 [s]	0.80 [s]	0.80 [s]	0.80 [s]
Minimum time lag	0.20 [s]	0.20 [s]	0.20 [s]	0.20 [s]	0.20 [s]	0.20 [s]	0.20 [s]
Maximum time lag	1.50 [s]	1.50 [s]	1.50 [s]	1.50 [s]	1.50 [s]	1.50 [s]	1.50 [s]

Figure 18. EddyPro file descriptions for input.

Basic Settings

1) Set the path to your raw data directory (this is the ./EP_Ready/ folder)
2) Input the raw file name format so EP knows the timestamp of your files
    a. An example file name from Jan 2nd, 2025, at 3:45am is:

"EP_Ready__PAW_20250102_0345.txt" to tell EP how to read the timestamp provide it with: "EP_Ready__PAW_yyyymmdd_HHMM.txt"

3) Provide path to the folder you would like the output saved to and an output ID. Naming the ID according to the site and bins which are calculated is a good idea, i.e. PAW_123

4) Under the variables tab select CO_2, H_2O, and CH_4 for the first three tabs (also select N_2O for the 4th tab if you are also calculating total fluxes)

Advanced Settings

Advanced settings has four sections: processing options, statistical analysis, spectral analysis and corrections, and output files. Below are the settings to be applied for each section, **Table 3, 4, 5, 6**. If a setting is not explicitly mentioned in the lists below, then it should be left as the default.

Table 3. Processing options settings for EddyPro.

Wind speed measurement offsets	0.00 m/s for all (U,V,W)
Fix 'w boost' bug	Uncheck
Angle-of-attack correction	Uncheck
Axis rotations for tilt correction	Check, double rotation
Turbulent fluctuations	Block average
Time lags compensation	Check, Constant
Compensate density fluctuations	Uncheck
Add instrument sensible heat components	Uncheck
Quality check	Check, Mauder and Foken (2004)
Footprint estimation	Check, Kljun et al. (2004)

Table 4. Statistical analysis settings for EddyPro.

Spike count/removal	Check, and change CO2, H2O, CH4, and 4th Gas to 20.0 σ
Absolute limits	Check, and change the following: Filter outranged values, uncheck
ALL other tests	Uncheck
Random uncertainty estimation	Check, and change the following: Max correlation period, 50.0 seconds

Table 5. Spectral analysis and correction settings for EddyPro.

Filter (co)spectra according to...	Uncheck
Low data quality	Uncheck
Moderate data quality	Uncheck
Analytic correction of high-pass filtering...	Uncheck
Correction of low-pass filtering effects	Uncheck
Correction for instrument separation	Check, (2009) only crosswind and vertical

Table 6. Output file settings for EddyPro.

Full Output	Check
Use Fluxnet standard...	Check
Set error label...	Check
Biomet measurements	Uncheck
Details of steady state...	Uncheck
Metadata	Check
Output format	Output only available results
Build continuous dataset	Check
Reduced spectra and ogives	Check ALL
Full length spectra	Uncheck ALL
Full length cospectra	Uncheck ALL
Statistics	Check Level 1 and 2
Time series	Check Level 1 and 2
Variables	Check ALL (except uncheck Tair and Pair)

Run Output

With all these settings applied, run EP in Advanced mode. If you only run this in express mode only your basic settings will be applied.

Warnings and errors may appear. “Warning (53) No raw data file relevant to current averaging period was found” is generally the only warning/error that is acceptable (it is by EP reaching a time with a gap in data, so expect it whenever you have gaps). Other warnings/errors should be read carefully. Often, errors which stop EP from starting are often caused by a mismatch between your files and the file description provided in Project Creation.

Contents of EddyPro Output

The structure of the output produced by EddyPro depends on the metadata file used and the advanced settings options. If you applied different settings than what is listed above your output files may look different. Several CSV files, and folders containing more CSVs, will be included in the output. The file containing fluxes for the processed bins is named after whatever the project was titled as but includes "_full_output_" and is also a CSV. Data begins on the 4th row; each row represents a 30-minute period. The 1st, 2nd, and 3rd rows contain descriptions of the data, column headers, and units, respectively.

Fluxes for each 30 minute period are found in the columns: co2_flux, h2o_flux, ch4_flux, and n2o_flux. If metadata files are applied correctly co2_flux will contain bin 1,4, and 7 fluxes. Bins 2, 5, and 8 are under h2o_flux. Bins 3, 6, and 9 are under ch4_flux. The total aerosol flux should be under n2o_flux only.

Before moving on with your data open the "full_output" csv in Excel to see any errors (-9999 values) readily. It's recommended to check that you don't see error values for any of the times you have POPS and anemometer data in your "EP_Ready" folder. If you do see errors at times you don't expect re-run EddyPro and ensure you provided correct information in the Basic Settings tab.