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# Identifying Better Indicators of Aerosol Wet Scavenging During Long-Range Transport

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**Abstract.** As the dominant sink of aerosol particles, wet scavenging greatly influences aerosol lifetime and interactions with clouds, precipitation, and radiation. However, wet scavenging remains highly uncertain in models, hindering accurate predictions of aerosol spatiotemporal distributions and downstream interactions. In this study, we present a flexible, computationally inexpensive method to identify meteorological variables relevant to estimating wet scavenging using a combination of aircraft, satellite, and reanalysis data augmented by trajectory modeling to account for air mass history. Treating the enhancement (Δ) ratio of black carbon and carbon monoxide (ΔBC/ΔCO) measured by aircraft as an in situ proxy for wet scavenging, we assess the capabilities of an array of meteorological variables to predict ΔBC/ΔCO using regression statistics derived from curve-fitting and k-fold cross-validation. We find that accumulated precipitation along trajectories (APT) – treated as a wet scavenging indicator across multiple studies – is unable to accurately capture ΔBC/ΔCO trends, suggesting that APT is not a good indicator of wet scavenging effects. In contrast, the frequencies of precipitation or high relative humidity along trajectories better predict ΔBC/ΔCO trends and magnitudes, suggesting that these types of meteorological variables are better than APT for estimating the degree of wet scavenging in an air mass. Precipitation characteristics (e.g., intensity, frequency) from satellite retrievals are better indicators of ΔBC/ΔCO than those calculated from reanalysis, supporting previous studies that demonstrated reanalysis to be less reliable than satellite retrievals in terms of precipitation. Finally, top quantiles (e.g., 90$^{th}$) of relative humidity are able to consistently capture the behavior of ΔBC/ΔCO and may also be a more suitable indicator of wet scavenging than APT. Future studies can use the best-performing meteorological variables identified in our study to estimate wet scavenging. Furthermore, this method can be repeated for different regions to identify region-specific factors influencing wet scavenging, and our findings may be useful for informing scavenging parametrization schemes in models.

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## 1 Introduction

Although wet scavenging is the dominant removal mechanism for atmospheric aerosol particles (Seinfeld and Pandis, 2016; Textor et al., 2006), it remains a large source of uncertainty in global-scale models (Watson-Parris et al., 2019; Liu and Matsui, 2021; Moteki et al., 2019; Hodzic et al., 2016). This uncertainty hampers the ability of global-scale models to capture the lifecycle (i.e., sources, transformations, and sinks) (Hou et al., 2018), spatial extent (Moteki et al., 2019), and vertical profile (Watson-Parris et al., 2019; Liu and Matsui, 2021; Frey et al., 2021; Kipling et al., 2016) of aerosol particles. Inaccurate representations of these aerosol features contribute to uncertainties in estimates of aerosol radiative effects (Samset et al., 2013; Marinescu et al., 2017) and aerosol loadings over climate-sensitive regions (Liu and Matsui, 2021; Mahmood et al., 2016; Shen et al., 2017), with further implications for the remote sensing of aerosol abundance downwind of precipitating or cloudy areas. Advancing knowledge of wet scavenging processes can help reduce the largest uncertainty in human forcing of the climate system, which involves aerosol-cloud interactions (e.g., Bellouin et al., 2020).

Wet scavenging occurs either below- or in-cloud. Below-cloud scavenging occurs when aerosol particles are collected by precipitation (Croft et al., 2009) and is most important between the surface and 1 km above ground level (AGL) (Grythe et al., 2017). The efficiency of below-cloud scavenging depends on raindrop size distributions and aerosol composition (Lu and Fung, 2018; Grythe et al., 2017) as well as the amount of in-cloud condensed water (Luo et al., 2019). To calculate the fraction of aerosol scavenged below-cloud, models typically rely on an empirically-derived below-cloud scavenging coefficient, which is a function of aerosol size (Feng, 2007; Croft et al., 2009) and composition (Lin et al., 2021). Semi-empirical model parametrizations of below-cloud scavenging have been shown to improve simulated surface concentrations (Luo et al., 2019); however, agreement between models and observations is highly sensitive to the specific below-cloud scavenging scheme used (Lu and Fung, 2018). Below-cloud scavenging rates in models also remain significantly underestimated compared to observations (Kim et al., 2021; Ryu and Min, 2022; Xu et al., 2019).

In-cloud scavenging occurs via nucleation (i.e., activation of aerosol particles into cloud droplets; Jensen and Charlson, 1984) or impaction (i.e., collision of interstitial aerosol particles with existing cloud droplets; Kipling et al., 2016; Flossmann et al., 1985) and is followed either by (1) precipitation that reaches the surface, removing the particle from the atmosphere (Radke et al., 1980), or (2) evaporation of cloud droplets or precipitation, returning the scavenged particle to the free atmosphere (Mitra et al., 1992). Model improvements in in-cloud scavenging include using a continuous rather than binary cloud fraction (Ryu and Min, 2022; Xu and Randall, 1996), accounting for cloud water phase (Grythe et al., 2017; Liu and Matsui, 2021), and accurately simulating cloud supersaturation (Moteki et al., 2019). Although in-cloud scavenging is generally thought to be more efficient at removing accumulation mode aerosol particles (Watson-Parris et al., 2019; Choi et al., 2020), other studies argue that there are instances wherein below-cloud scavenging becomes more important at regulating aerosol burdens (Kim et al., 2021; Ryu and Min, 2022; Xu et al., 2019). Uncertainties related to wet scavenging are further exacerbated by the divergent role of clouds, which can be a sink or source of aerosol particles depending on environmental factors and cloud characteristics (Ryu et al., 2022).

One avenue for improving the estimation of wet scavenging, particularly in observational studies, is to identify an effective indicator of wet scavenging. Previous studies used precipitation amount (Feng, 2007; Andronache, 2003) while more recent studies accounted for air mass history using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory Model (Rolph et al., 2017; Stein et al., 2015) to calculate accumulated precipitation along trajectories (APT) (e.g., Kanaya et al., 2016, 2020). However, APT can be problematic as an indicator of wet scavenging because APT is an accumulated quantity and does not consider specific characteristics of precipitation relevant to scavenging such as intensity and frequency (Hou et al., 2018; Wang et al., 2021c, b; Hilario et al., 2022). APT as an indicator of wet scavenging also

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relies on the correct detection of precipitation and retrievals of amounts, which are challenging during both light (Nadeem et al., 2022; Kidd et al., 2021) and intense precipitation events (Chen et al., 2020a; Gupta et al., 2020) and even show disagreements between different satellite precipitation products (SPPs) and reanalyses (Cannon et al., 2017; Jiang et al., 2021; Alexander et al., 2020; Chen et al., 2020b; Barrett et al., 2020). Furthermore, precipitation from SPPs such as the Precipitation Estimation from Remotely Sensed Information using Artificial Neural Networks – Climate Data Record (PERSIANN-CDR) (Ashouri et al., 2015; Nguyen et al., 2018) and the Integrated Multi-satellitE Retrievals for the Global Precipitation Measurement (GPM) mission (IMERG) (Huffman et al., 2020) refer to total column precipitation that have been validated mainly with surface measurements (Sapiano and Arkin, 2009; Nicholson et al., 2019; Wang et al., 2021a) and consequently may not detect precipitation that evaporates before reaching the surface (e.g., virga) (Wang et al., 2018).

Given the uncertainties of estimating wet scavenging from precipitation, we present a flexible, computationally inexpensive method to identify alternative meteorological variables that can be used to better estimate wet scavenging. We combine curve-fitting and k-fold cross-validation to evaluate an array of meteorological variables from aircraft, satellite, and reanalysis data to answer the following:

(1) What meteorological variables can estimate wet scavenging trends better than APT? Since precipitation frequency has been shown to exert significant control over aerosol scavenging (Wang et al., 2021b), we hypothesize that predictors that account for the frequency of scavenging-conducive conditions (e.g., frequency of high relative humidity (RH) conditions along trajectories) will be able to capture wet scavenging trends better than APT.
(2) How can APT be filtered or changed to better estimate wet scavenging? We hypothesize that considering precipitation intensity and/or trajectory altitude thresholds when calculating APT will improve its ability to estimate wet scavenging. We also hypothesize that calculating APT using SPPs will perform better than APT from reanalysis.

The presented method may be repeated over different regions to identify region-specific wet scavenging indicators. This can inform scavenging parameterization development for models by providing guidance on what meteorological variables are needed to properly capture wet scavenging processes over a specific region. Future studies can also use the best-performing variables identified in this study as alternatives to APT when estimating the extent of wet scavenging.

## 2 Data & Methods

### 2.1 Aircraft data

Much of the methodology and instrument details in this study are detailed elsewhere (Hilario et al., 2021) but are summarized here. We utilize aircraft measurements from NASA's Cloud, Aerosol, and Monsoon Processes-Philippines Experiment (CAMP$^2$Ex; 24 August to 5 October 2019) over the tropical West Pacific (Reid et al., 2023), which hosts a dynamic transport environment rich in aerosol sources and cloud-precipitation systems.

Black carbon (BC)-equivalent concentrations (particle diameters: 100 – 700 nm) were measured with a Single-Particle Soot Photometer (SP2) (Moteki & Kondo, 2007, 2010) with an uncertainty of 15% (Slowik et al., 2007) and lower detection limit of 10 ng m$^{-3}$ verified by filter-blank measurements as well as observations in the clean free troposphere. To eliminate in-cloud sampling artifacts such as droplet shattering on the inlet (Murphy et al., 2004), we use only data collected outside of clouds. All BC concentrations are reported at standard temperature and pressure (273 K, 1013 hPa). Carbon monoxide (ppm) was measured using a dried-airstream near-infrared cavity ringdown absorption spectrometer (G2401-m; PICARRO, Inc.), with an uncertainty of 2% and precision of 0.005 ppm. As an in situ (i.e., at the aircraft's position) contrast to moisture-based variables along trajectories,

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relative humidity ($RH_{W, DLH}$) was derived from absolute water vapor concentrations that were retrieved by a diode laser hygrometer (DLH) (Livingston et al., 2008) on the aircraft.

### 2.2 Enhancement ratio calculation

To relate wet scavenging to transport conditions, previous studies used enhancement (Δ) ratios of BC and carbon monoxide (CO) (ΔBC/ΔCO; Hilario et al., 2021; Kanaya et al., 2016; Oshima et al., 2012). By using the enhancement above background, ΔBC/ΔCO accounts for spatial variations in the background levels of BC and CO and is better able to detect an air mass containing BC and CO above background levels. This ratio can be used as an indicator of wet scavenging because BC is relatively chemically inert and is mainly removed from the atmosphere via wet scavenging (Moteki et al., 2012). While CO is also relatively chemically inert, CO has a lifetime between 30 to 90 days (Seinfeld and Pandis, 2016) that is mainly controlled by photochemistry rather than wet scavenging due to its low solubility. Because of this, we use ΔBC/ΔCO as our proxy for wet scavenging and the predictand for our regression models.

Enhancements were defined as the difference between species concentrations and the lowest 5$^{th}$ percentile species concentration for all CAMP$^2$Ex data for every 5 K potential temperature bin (Koike et al., 2003; Matsui et al., 2011). As CAMP$^2$Ex spanned the late southwest monsoon and early monsoon transition, background concentrations (i.e., lowest 5$^{th}$ percentile) were calculated for each monsoon phase using 20 September 2019 to divide the two monsoon phases. Only data with ΔCO > 0.02 ppm were included to reduce uncertainties caused by low denominator values in the ΔBC/ΔCO ratio (Kleinman et al., 2007; Kondo et al., 2011; Matsui et al., 2011).

### 2.3 Trajectory modeling

Trajectory modeling is a computationally inexpensive tool for characterizing transport processes (Kanaya et al., 2016; Oshima et al., 2012; Moteki et al., 2012) and has been used in synergy with aircraft data (Hilario et al., 2021; Dadashazar et al., 2021). In this study, we use trajectories to account for meteorological conditions during air parcel transport that are expected to impact the scavenged aerosol fraction. Backward trajectories were spawned every minute along the aircraft flight path and run for 72 hours using the NOAA HYSPLIT model. Meteorological input data for the HYSPLIT model were from the National Centers for Environmental Prediction (NCEP) Global Forecast System reanalysis (GFS; 0.25° × 0.25°).

### 2.4 Data for predictor variables

Several meteorological variables (i.e., predictors) considered in this work were calculated from GFS reanalysis collocated along each trajectory. Though reanalysis is relatively coarse and not cloud-resolving, reanalysis variables (e.g., RH) may still be useful in detecting the presence of meso-to-synoptic-scale cloud fields. As precipitation is expected to be accompanied by elevated RH or water vapor mixing ratio (MR), these reanalysis-derived variables could serve as effective scavenging indicators in cases where precipitation may be missed or misestimated.

In addition to APT from GFS, we calculated APT from two SPPs: PERSIANN-CDR (0.25° × 0.25°, daily resolution) (Ashouri et al., 2015; Nguyen et al., 2018) and IMERG Final v6 (0.1° × 0.1°, 30-min resolution) (Huffman et al., 2020). We converted precipitation from these products to hourly amounts to match trajectory timesteps prior to further calculation.

Besides APT, we also calculated precipitation amount (PA; mm $h^{-1}$), frequency (PF), and intensity (PI; mm $h^{-1}$), which are well-established in the literature for characterizing precipitation, particularly in diurnal cycle analyses (e.g., Zhang et al., 2017; Hilario et al., 2020). Applying these quantities to precipitation along trajectories, PA is APT divided by the total number of hours along the trajectory (i.e., trajectory length) to obtain an average hourly precipitation rate, PF is the fraction of hours along the

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trajectory where the grid cell precipitation is above 0 mm, and PI is the ratio of PA to PF. Table 1 shows notation used to explain each type of predictor and its variations.

### 2.5 Curve-fitting and k-fold cross-validation

To quantify relationships between ΔBC/ΔCO and each predictor, we performed k-fold cross-validation (k = 10) parallelized using the Python package jug (Coelho, 2017). To create k distinct partitions of the data, we utilized stratified random sampling wherein random sampling was performed for every 5$^{th}$ percentile block of the predictor such that the sampling probability better reflects the distribution of predictor values (e.g., precipitation) and the resulting partition captures the behavior of ΔBC/ΔCO over the whole range of predictor values. By randomly sampling each percentile block for k distinct partitions, this sampling method improves the chances of capturing intra-block variability in ΔBC/ΔCO by collecting the most samples where the most data exist. The random nature of the sampling also allows for the consideration of extreme values in the curve-fitting, with a sampling probability proportional to the frequency of these extreme values. As an example, Fig. 1a shows the emphasis of the stratified random sampling method on high density areas of the scatterplot of $RH_{q90}$ and ΔBC/ΔCO, denoted by dense percentile blocks. For extremely skewed distributions such as APT, several of the lower-value percentiles exhibited non-unique values (e.g., zero). In the case of repeated percentile values, these percentile-based groups were merged. We imposed a minimum of six distinct percentile blocks to ensure robust curve-fitting.

An iterative train-test split procedure using these partitions was then performed using nine partitions as the training set and the remaining partition as the testing set (Fig. 1a). For each iteration of the k-fold cross-validation, nonlinear least squares curve-fitting was applied to the training set (i.e., 9 partitions), to determine coefficients for the equation (e.g., general exponential; discussed below) fitted onto the scatterplot of ΔBC/ΔCO and the predictor. We used these coefficients and the testing set (i.e., the remaining partition) as inputs for the curve-fitting equation and calculated a predicted curve of ΔBC/ΔCO and the predictor. To assess this predicted curve, we applied stratified random sampling on the testing set and took the median ΔBC/ΔCO per 5$^{th}$ percentile block to create observed curves of ΔBC/ΔCO as a function of the predictor that could be compared to the predicted curve (Fig. 1b). Because decreases in ΔBC/ΔCO are expected to be mainly from wet scavenging, the overall trend or median curve may be treated as a reasonable indicator of wet scavenging effects on ΔBC/ΔCO related to changes in the predictor value.

Using a linear regression of predicted and observed median ΔBC/ΔCO per 5th percentile block of the predictor (Fig. 1c), we calculated statistics to describe the performance of the predictor (e.g., slope, R). Note that the performance of a predictor refers to how well ΔBC/ΔCO derived from the predictor matches observed ΔBC/ΔCO. We also computed statistics comparing predicted and observed ΔBC/ΔCO for individual points (Fig. 1d) rather than medians to assess how much variability in ΔBC/ΔCO is captured by the predicted curves. These individual-point statistics resulted in correlations and slopes further from ideal values compared to the median-based statistics. This is expected as individual ΔBC/ΔCO points have high variability due to factors other than wet scavenging; however, a comparison of individual-point and median-based statistics show that they qualitatively agree quite well, with the relative ranking of predictors largely unchanged between the two types of statistics. In other words, the top predictors performed well whether we used median-based or individual-point statistics, implying the conclusions reached using our method are qualitatively unaffected. For simplicity, reported statistics in this study refer to median-based statistics unless otherwise specified.

To determine if a predicted ΔBC/ΔCO curve tended to overestimate or underestimate observed ΔBC/ΔCO, we calculated a weighted area difference (WAD) using Eq. 1:

$$WAD = \frac{\sum N_i \cdot x_i}{\sum N_i} \quad (1)$$

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where $x_i$ is the difference between observed and predicted ΔBC/ΔCO for the $i^{th}$ percentile block and $N_i$ is the number of data points in that percentile block. A positive (negative) WAD indicates an overestimate (underestimate) of observed ΔBC/ΔCO.

To account for differing relationships between ΔBC/ΔCO and each predictor, we fitted their scatterplot using multiple nonlinear equations (Table 2) and chose the equation producing the highest Pearson correlation (R) between observed and predicted ΔBC/ΔCO for that predictor. We use R because we are more interested in predictors that can capture trends in ΔBC/ΔCO rather than magnitudes. For some combinations of predictors and curve-fitting equations, the curve-fitting did not successfully converge (< 4% of all combinations and k-fold iterations). In these cases, we did not include the predictor-equation combination in our analysis. However, curve-fitting on the predictor may still converge when using a different equation. In such a scenario, the predictor becomes part of our analysis.

This procedure was repeated for k iterations using a different partition for the testing set in each iteration, which provides a measure of uncertainty in the resulting regression statistics. Sensitivity testing with the *k* value showed no significant effect on the general conclusions of the study.

Although there is no physical process built into this procedure, the strength of the method is its repeatability in different environments or regions with minimal changes to the overall procedure. As it requires no physical model to be run besides the trajectory calculations, the method is also relatively computationally inexpensive. Future work wanting a more physical basis may apply our method as a diagnostic tool to identify and narrow down a list of meteorological variables that may be relevant to wet scavenging and continue their analysis with a physical model using the narrowed list of variables to analyze.

## 3 Results and Discussion

### 3.1 Overall statistical performance

Figures 2-3 show performance comparisons of different predictors derived from linear regressions of observed and predicted ΔBC/ΔCO. Hereafter, the performance of a predictor in this study refers to a predictor's ability to reproduce observed ΔBC/ΔCO via curve-fitting (Sect. 2.5; Fig. 1). A threshold of Pearson R > 0.71 ($R^2$ > 0.50) was used to narrow our analysis to predictors that were able to produce predicted ΔBC/ΔCO that captured trends in observed ΔBC/ΔCO. We note that no APT-related predictor met this R threshold, suggesting that APT may not be a good predictor of ΔBC/ΔCO and, by extension, aerosol scavenging. One explanation is that APT does not account for different precipitation characteristics such as precipitation frequency or intensity, both of which have been argued to be important for regulating aerosol scavenging (Hou et al., 2018; Wang et al., 2021c). To explore this possibility further, we calculated PA, PF, and PI for each trajectory (Sect. 2.4). Among these three, PF resulted in the most predictors satisfying the minimum R of 0.71. Several PF-related variables in Fig. 2 have R over 0.71, even when accounting for the 25$^{th}$ and 75$^{th}$ percentile error bars derived from k-fold cross-validation. In comparison, only one PA-related predictor ($PA_{48H, IMERG}$; Fig. 2a) and one PI-related predictor ($PI_{PCP > 0.2mm, 48H, P-CDR}$; Fig. 2b) pass the R > 0.71 threshold, which indicates that ΔBC/ΔCO predicted by PA- or PI-related variables do not track observed ΔBC/ΔCO as well as PF-predicted ΔBC/ΔCO. These results suggest that PF may be more important than PA and PI for predicting aerosol scavenging over the tropical West Pacific.

Noticeably, only one precipitation-related predictor calculated from GFS met the R threshold ($PI_{PCP > 0.mm2, 72H, GFS}$; Fig. 2b). This suggests that GFS-derived precipitation variables are not able to capture observed ΔBC/ΔCO trends in contrast to several SPP-based precipitation variables that showed moderate to strong R (> 0.71) with ΔBC/ΔCO. The poor performance of GFS-derived precipitation is reflective of past studies showing disagreements in precipitation characteristics between satellite and reanalyses (Cannon et al., 2017; Jiang et al., 2021) and even divergent precipitation trends and amounts among individual reanalysis products (Alexander et al., 2020; Chen et al., 2020b; Barrett et al., 2020). Our results suggest that precipitation from GFS reanalysis

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is not a reliable predictor of aerosol scavenging compared to precipitation from SPPs. Future studies relating precipitation to aerosol scavenging are recommended to instead rely on in situ or satellite retrieved precipitation rather than precipitation from reanalysis.

Predictors based on quantiles of RH (e.g., $RH_{q90}$) perform quite well based on R, slope (Fig. 2e), intercept, and WAD (Fig. 2e). $RH_{q90}$ performs slightly better than other RH thresholds (Fig. 2e); however, this difference is minor as shown by the overlapping $25^{th}$-$75^{th}$ percentile error bars between the different RH thresholds. The similar performance between different RH quantiles suggests consistency in their ability to predict ΔBC/ΔCO trends (high R) while doing reasonably better than other types of predictors when estimating ΔBC/ΔCO magnitudes across the spectrum of predictor values (intercepts closer to 0, slopes closer to 1). Maximum RH along trajectories was used by Kanaya et al. (2016) in their analysis of ΔBC/ΔCO scavenging to detect the role of clouds in BC removal. Our findings suggest that top quantiles of RH, including its maximum, are a good choice for predicting scavenging trends.

Compared to variables directly linked to precipitation (PA, PF, PI, APT), the slopes from RH quantiles are noticeably closer to the ideal value of 1 (Fig. 2e) while intercepts from RH quantiles are closer to the ideal value of 0 (Fig. 2e), meaning ΔBC/ΔCO predicted by RH quantiles more closely matches the magnitude of observed ΔBC/ΔCO compared to ΔBC/ΔCO predicted by precipitation. We hypothesize that the better performance of RH-related predictors over those more directly related to precipitation (e.g., APT) may be explained by instances of precipitation that is missed (or misestimated) by SPP retrievals that is indirectly detected by reanalysis as high humidity conditions. This possibility is supported by previous literature showing the tendency of SPPs to misestimate light (Nadeem et al., 2022; Kidd et al., 2021) or intense precipitation (Chen et al., 2020a; Gupta et al., 2020); however, our hypothesis of the connection between RH from reanalysis and precipitation from SPPs requires further investigation in future work.

Of all the fractional predictors considered in this study, $f_{MR15}$ and $f_{RH95}$ perform the best (Fig. 2d). Interestingly, the $25^{th}$-$75^{th}$ percentile range of slopes from $f_{RH95}$ (0.81 – 1.20; Fig. 2d) overlaps with the ideal value of 1 and its intercepts (~0.25; Fig. 2d) are lower than the other predictors in Fig. 2. These slope and intercept statistics indicate that $f_{RH95}$ is capable estimating the magnitude of ΔBC/ΔCO throughout the spectrum of $f_{RH95}$ values, meaning that $f_{RH95}$ can be used to capture ΔBC/ΔCO trends and magnitudes for both high-ΔBC/ΔCO (fresh) and low-ΔBC/ΔCO (scavenged) air masses. A similar performance is observed for $f_{MR15}$. $f_{MR15}$ and $f_{RH95}$ are similar to PF, which was the best-performing category of precipitation-related predictors, in that PF, $f_{MR15}$, and $f_{RH95}$ represent the occurrence frequencies of some condition along each trajectory (e.g., non-zero precipitation or RH > 95%). The better performance of these frequency-related predictors compared to other types of predictors suggests that the frequency of precipitation or high-RH conditions may be more reliable indicators of aerosol scavenging than the magnitude of precipitation (e.g., APT, PI, PA).

One limitation of our analysis is the goodness-of-fit achieved during the k-fold cross-validation process as the goodness-of-fit affects the validity of interpreting the resulting performance statistics (e.g., slope). Fig. S1 of the Supplementary Information (SI) shows no large difference in the goodness-of-fit between different predictors based on R and RMSE. Similar magnitudes of RMSE (Fig. S1) suggest that the interpretation of performance statistics derived from the curve-fitting procedure is equally valid across the discussed predictors.

### 3.2 Nonlinear sensitivity of ΔBC/ΔCO to meteorological variables

Although the predictors in Figs. 2 – 3 exhibit the highest R of all predictors considered in this study, their slopes are generally below 1 (Fig. 2) while their intercepts and WAD are generally positive (Fig. 3). The combination of these statistics implies that predictions of ΔBC/ΔCO using our method tend to overestimate observed ΔBC/ΔCO across the spectrum of predictor values (indicated by WAD > 0) with maximum overestimations occurring when observed ΔBC/ΔCO is low (indicated by slopes < 1 and

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intercepts > 0). This points to a nonlinear sensitivity of ΔBC/ΔCO to these predictors as the degree of scavenging increases. Dadashazar et al. (2021) observed a similar nonlinear response to APT by a ratio of particulate matter below 2.5 μm to CO ($\Delta PM_{2.5}/\Delta CO$), where $\Delta PM_{2.5}/\Delta CO$ was most responsive to APT when APT was below 5 mm and less sensitive to APT when APT exceeded 5 mm.

Investigating this sensitivity further, Fig. 4 shows that PF-predicted ΔBC/ΔCO does not capture the trends of observed ΔBC/ΔCO for highly scavenged air masses. In other words, PF loses its predictive power as the degree of scavenging increases, implying that PF is most important for the scavenging of fresher air masses (high-ΔBC/ΔCO). This nonlinear sensitivity of ΔBC/ΔCO to PF hints at the possibility that other meteorological variables may become important for further scavenging of highly scavenged air (low-ΔBC/ΔCO). In contrast to predictors directly related to precipitation (Fig. 4d-f), the predicted curves of $RH_{q95}$ (Fig. 4a), $f_{RH95}$ (Fig. 4b), and $f_{MR15}$ (Fig. 4c) visibly track the trends of observed ΔBC/ΔCO with approximately half the difference between predicted and observed ΔBC/ΔCO when ΔBC/ΔCO is low. The capability of $RH_{q95}$, $f_{RH95}$, and $f_{MR15}$ to predict ΔBC/ΔCO across a wider range of values is further reflected by generally lower intercepts and WAD (Fig. 3) than precipitation-related predictors, which suggests promising alternative indicators of aerosol scavenging. However, we also note that such differences could arise partly from the limitations of curve-fitting, wherein fitted curves naturally capture gradual changes (e.g., Fig. 4b) better than sharp ones (e.g., Fig. 4d).

### 3.3 Effect of filtering APT on performance

To improve the predictive performance of precipitation-related variables, we applied combinations of filters for (1) precipitation intensity, (2) trajectory altitude, (3) data product, and (4) trajectory length. Filtering for precipitation intensity isolates the contribution of higher precipitation intensities towards a predictor's ability to predict ΔBC/ΔCO. Intense precipitation has been shown to be more efficient at scavenging aerosol particles (Zhao et al., 2020) and may be important when estimating aerosol scavenging. Filtering for trajectory altitude (i.e., considering precipitation only when the trajectory altitude is below 1.5 km AGL) tests the hypothesis that air masses within the boundary layer will be most susceptible to wet scavenging. Grythe et al. (2017) demonstrated that below-cloud scavenging (i.e., impaction by precipitation) accounted for majority of scavenging events below 1 km. We selected 1.5 km based on previous work on the marine boundary layer over the tropical West Pacific (Chien et al., 2019). We repeated the analysis for three precipitation products (one reanalysis and two SPPs) to capture variability in our results due to the choice of data product which has been shown to be important for precipitation (Alexander et al., 2020). Finally, we tested the effect of trajectory length on the performance of APT as a predictor of ΔBC/ΔCO.

In general, we found that applying altitude and/or precipitation filters negatively affected the performance of APT (Fig. 5b-d), leading to lower R between predicted and observed ΔBC/ΔCO compared to the case without any filters applied (Fig. 5a). The poorer performance of precipitation filtered for higher intensities (Fig. 5b) suggests that these higher intensities may not be as important for estimating wet scavenging compared to low intensity precipitation, consistent with previous work showing that light rain exerts more control over the global aerosol burden (Wang et al., 2021b) and that precipitation over the tropical West Pacific is typically high in frequency and low to moderate in intensity (Biasutti et al., 2012).

Applying a filter for trajectory altitude prior to calculating APT also did not lead to improvements in R (Fig. 5c). This was surprising because, when using total column precipitation from SPPs, a maximum altitude filter should reduce errors from cases where precipitation occurs below the air mass and no scavenging occurs. Since the SPPs used in this study have been validated using surface measurements (Sapiano and Arkin, 2009; Nicholson et al., 2019; Wang et al., 2021a), precipitation from SPPs should be reflective of precipitation that reaches the surface, implying a susceptibility of these SPPs to errors related to virga (Wang et al., 2018). However, Wang et al. (2018) also showed that virga occurrence over the tropical West Pacific is also infrequent. An

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alternative explanation for the poor performance of altitude-filtered predictors is uncertainties related to trajectory altitude (Harris et al., 2005), such that an air parcel may have actually been traveling at a lower altitude than its modelled trajectory and underwent scavenging.

An examination of trajectory altitudes (Fig. S5) revealed that filtering for trajectory altitudes below 1.5 km excluded the majority (~70%) of precipitating grid cells encountered by trajectories, which likely negatively impacted the predictive ability of altitude-filtered predictors.

Longer trajectories resulted in slightly higher R between observed and APT-predicted ΔBC/ΔCO (Fig. 5a-b), increasing from R ~0.5 to ~0.6; however, this difference is not large, as shown by overlapping $25^{th}$-$75^{th}$ percentile error bars. Interestingly, this increase in R for longer trajectories was more evident when filtering for precipitation intensities > 0.2 mm $h^{-1}$ when calculating APT (Fig. 5b) or PI (Fig. S2b), but not when applying this intensity filter on PF (Fig. S3b) or PA (Fig. S4b). Further interpretation likely requires a physical model in future work to explain why the performance of intense precipitation (Fig. 5b) benefits from a longer trajectory more than total precipitation does (Fig. 5a).

## 4 Limitations

**ΔBC/ΔCO as a wet scavenging proxy**: In this study, we treat ΔBC/ΔCO as a proxy for wet scavenging (i.e., predictand) and base our conclusions on which variables best predict this ratio (i.e., predictors). An underlying assumption is that there is negligible emission of BC or CO after initial emission and after wet scavenging occurs. Consequently, this method is expected to work well in outflow regions such as the tropical West Pacific and not well where additional BC and/or CO are likely to be after initial emission or wet scavenging has occurred (e.g., continental region).

**ΔBC/ΔCO depends on air mass type**: The ΔBC/ΔCO quantity is also affected by air mass type. For example, biomass burning and anthropogenic/industrial emissions will have different ΔBC/ΔCO values (e.g., Hilario et al., 2021). The mixing state and composition of BC-containing particles will also affect its hygroscopicity and by extension its rate of wet scavenging (Liu et al., 2013). Although these factors are expected to influence ΔBC/ΔCO, we assume the response of ΔBC/ΔCO to the predictor will be chiefly determined by scavenging-related processes. While the response of ΔBC/ΔCO to APT in Hilario et al. (2021) was quite similar across different air mass origins (their Fig. 9), this may not be always the case. However, this origin-independent relationship does allow for the option of curve-fitting without differentiating between individual sources. Beneficially, this choice to aggregate the data rather than resolving by source increases the number of data points available for curve-fitting, which improves the robustness of the resulting statistics and adds strength to our conclusions on which meteorological variables are most relevant for aerosol scavenging.

**Specific processes:** The conclusions of our study are based on the relative ability of different variables to predict ΔBC/ΔCO, our proxy for wet scavenging. This approach does not isolate individual processes that are usually parameterized by global circulation models (e.g., impaction, nucleation) (Croft et al., 2009, 2010; Ryu and Min, 2022). However, through our proposed framework, we can still gain qualitative insights into which meteorological variables are relevant for estimating aerosol scavenging, which can inform future studies as well as developments in model parametrization.

**Single predictor method:** The method presented here assesses the one-to-one relationship between a single predictor and ΔBC/ΔCO repeated individually for several predictors. We expect that using a combination of predictors may lead to better predictions of ΔBC/ΔCO while providing a more physical picture of relative contributions of different meteorological variables towards wet scavenging. Future work may utilize multiple linear regression or more sophisticated methods such as machine learning to consider different combinations of predictors with the objective of identifying a combination that predicts ΔBC/ΔCO

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well and extracting further information on what physical mechanisms may be relevant for the removal of ΔBC/ΔCO based on relative coefficients or weightings of different predictors.

**Curve-fitting**: The results can depend on the curve-fitting function used. Different variables are expected to have different relationships with ΔBC/ΔCO. Thus, if we considered only one function for curve-fitting, it would favor variables that have a specific relationship with ΔBC/ΔCO. To reduce this bias, we applied four different curve-fitting functions on each predictor based on previous studies (Table 2) that accounted for possible relationships between ΔBC/ΔCO and each predictor. We then chose the curve-fitting function that produced the highest R between observed and predicted ΔBC/ΔCO. However, we note that this does not completely remove the bias as specific functions were still selected.

**Trajectory modeling:** Vertical motion through convection, entrainment, and detrainment processes are known uncertainties in trajectory modeling, which increase with trajectory length (Harris et al., 2005). The spatial and temporal resolutions of the meteorological input used for the HYSPLIT model are also limiting factors as meteorology along HYSPLIT trajectories do not account for sub-timestep or sub-grid processes.

## 5 Conclusions

We present a method to identify meteorological indicators of aerosol scavenging using a combination of aircraft, satellite, and reanalysis data coupled with HYSPLIT backward trajectories. We apply this method to the CAMP$^2$Ex field campaign over the tropical West Pacific, which hosts a wide range of cloud fractions and precipitation characteristics as well as an environment characterized by long-range transport of aerosol and trace gas species. Since ΔBC/ΔCO is mainly affected by scavenging, we treat ΔBC/ΔCO as an in situ proxy for aerosol scavenging and evaluate the responses of ΔBC/ΔCO to different meteorological variables (i.e., predictors). The main conclusions of the study are the following:

1. Although APT has been utilized in several studies as an indicator of aerosol scavenging, we demonstrated that APT-predicted ΔBC/ΔCO does not track observed ΔBC/ΔCO well. Furthermore, the application of altitude or precipitation intensity filters negatively impacted the performance of APT in predicting ΔBC/ΔCO trends. Since APT is an accumulated precipitation amount over the whole trajectory, APT does not account for other precipitation characteristics such as intensity or frequency, which have been shown to be relevant for aerosol scavenging. This shortcoming may explain the overall poor relative performance of APT in predicting ΔBC/ΔCO.
2. To investigate which precipitation characteristics are most relevant for predicting aerosol scavenging, we calculated PA, PF, and PI along trajectories using precipitation from reanalysis (GFS) and SPPs (IMERG, PERSIANN-CDR). While several precipitation-related predictors calculated from SPPs were able to predict ΔBC/ΔCO reasonably well, only one precipitation-related predictor from GFS correlated with ΔBC/ΔCO ($R > 0.71$), suggesting that precipitation from SPPs may be better at predicting aerosol scavenging than precipitation from reanalysis. The poorer performance of precipitation from reanalysis in predicting ΔBC/ΔCO than from SPPs is corroborated by previous studies that found larger misestimates of precipitation by reanalysis than by SPPs. Because of our results and those of past studies, we recommend relying on in situ or SPP precipitation rather than precipitation from reanalysis, particularly when relating precipitation to aerosol scavenging.
3. Frequency-related predictors such as PF, $f_{MR15}$, and $f_{RH95}$ performed much better than APT in predicting ΔBC/ΔCO trends and magnitudes. PF, $f_{MR15}$, and $f_{RH95}$ represent the frequencies of occurrence of some meteorological condition along trajectories such as non-zero precipitation (PF) or RH exceeding 95% ($f_{RH95}$). The relatively better performance of these

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frequency-related predictors over those related to PA or PI suggests that the frequency of precipitation or high-RH conditions may be a more reliable indicator of aerosol scavenging than the magnitude of precipitation.

4. Predictors based on specific quantiles or the mean of RH also performed quite well in predicting both ΔBC/ΔCO trends and magnitudes. We found only minor differences in the performance depending on the exact quantile used. We hypothesize the outperformance of RH quantiles over predictors more directly related to precipitation (e.g., APT) to be due to missed precipitation in SPP retrievals that was indirectly represented in reanalysis as high humidity conditions. Such a possibility is supported by previous literature demonstrating that SPPs tend to misestimate light precipitation (Nadeem et al., 2022; Kidd et al., 2021); however, further work is required to explore this hypothesis.

By identifying which meteorological variables are relevant for predicting ΔBC/ΔCO trends (and by extension, aerosol wet scavenging), the findings of this study may be useful for informing future aerosol scavenging studies. Furthermore, these results may also aid in improving scavenging parametrization schemes in models. The method presented in this study is repeatable over a variety of environments and is relatively computationally inexpensive to apply. Future work may utilize machine learning to identify combinations of meteorological variables to predict aerosol scavenging and apply our method to different regions to identify region-specific indicators of aerosol scavenging. Furthermore, CAMP$^2$Ex included a rich dataset on cloud water composition (Crosbie et al., 2022; Stahl et al., 2021) that can be explored, as in past work for other regions (MacDonald et al., 2018), to gain additional insights into aerosol wet scavenging processes.

*Code availability.* Codes are freely available upon request to the authors.

*Data availability.* All datasets are publicly available and accessible at https://doi.org/10.5067/Suborbital/CAMP2EX2018/DATA001 (NASA/LaRC/SD/ASDC, 2020). HYSPLIT data are accessible through the NOAA READY website (https://www.ready.noaa.gov/index.php, last access: 11 April 2023) (NOAA Physical Sciences Laboratory, 2020).

*Author contributions.* EC, LDZ, MAS, JPD, GSD contributed to data collection. MRAH and AS conceptualized the study. MRAH performed the data analysis and prepared the manuscript with input from all co-authors.

*Competing interests.* The authors declare that they have no conflict of interest.

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**Table 1: Examples of notation used in this study.**

| Predictor | Description | Variations |
|---|---|---|
| $f_{RH95}$ | Fraction of hours along trajectories where GFS relative humidity (RH) > 95% | $f_{RH85}$, $f_{RH90}$ |
| $f_{MR15}$ | Fraction of hours along trajectories where GFS water vapor mixing ratio (MR) > 15 g $kg^{-1}$ dry air | $f_{MR17}$ |
| $RH_{q95}$ | 95$^{th}$ percentile of RH along trajectories | $RH_{q50}$, $RH_{q85}$, $RH_{q90}$, $RH_{q100}$, $RH_{mean}$ |
| $RH_{w,\ DLH}$ | RH over water measured by DLH onboard the aircraft | - |
| $APT_{PCP\ >\ 0.2\ mm,\ 48H,\ GFS,\ <\ 1500m}$ | Accumulated precipitation calculated along 48-h trajectories where GFS precipitation is above 0.2 mm and trajectory altitude is below 1500 m | Trajectory duration: 12H, 24H, 48H, 72H<br>Precipitation product: GFS, IMERG, PERSIANN-CDR<br>Maximum altitude filter: no filter, < 1500 m<br>Minimum precipitation filter: no filter, > 0.2 mm<br>Other precipitation variables: PA, PF, PI |

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**Table 2: Curve-fitting equations considered where x is the predictor variable and y is the observed ΔBC/ΔCO while a, b, c, and d are best-fit parameters determined via least-squares regression.**

| Name | Equation | Source |
|---|---|---|
| Gaussian | $y = a \cdot \exp(-\frac{(x-b)^2}{2 \cdot c^2}) + d$ | - |
| General Exponential | $y = a \cdot exp(-b \cdot x) + c$ | - |
| Oshima | $y = b - a \cdot \log_{10}(x)$ | Oshima et al. (2012) |
| Kanaya | $y = c \cdot exp(-a \cdot x^b)$ | Kanaya et al. (2016) |

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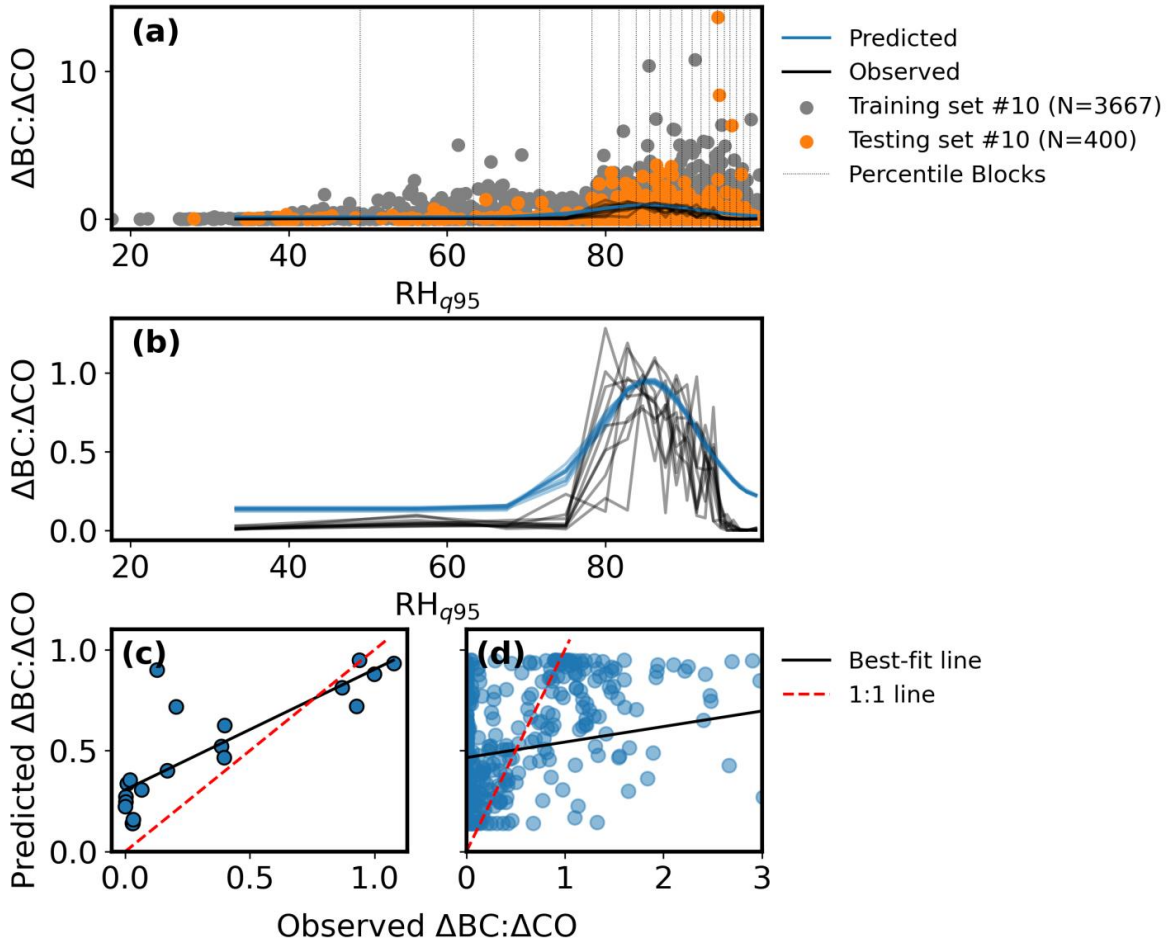


**Figure 1: An example of the curve-fitting procedure on ΔBC/ΔCO with $RH_{q95}$ as the predictor fitted with a gaussian function. (a) Training (gray dots) and testing sets (orange dots) for the 10th iteration of the k-fold cross-validation procedure selected using stratified random sampling. Percentile blocks are denoted by vertical gray lines with observed (black) and predicted curves (blue) also plotted for all 10 iterations. (b) Same as (a) but only showing observed (black) and predicted curves (blue) for all 10 iterations to highlight variations between the k iterations. (c) Scatterplot comparing $RH_{q95}$-predicted ΔBC/ΔCO and observed median ΔBC/ΔCO per 5th percentile block of the predictor. Note that (c) is simply the linear regression of the observed and predicted curves in (b). (d) Same as (c) but comparing $RH_{q95}$-predicted and observed ΔBC/ΔCO for individual points. In (c-d), only training set data are used, Y-axes are the same, the best-fit line is shown as a black line, and the 1:1 line is the red, dashed line.**

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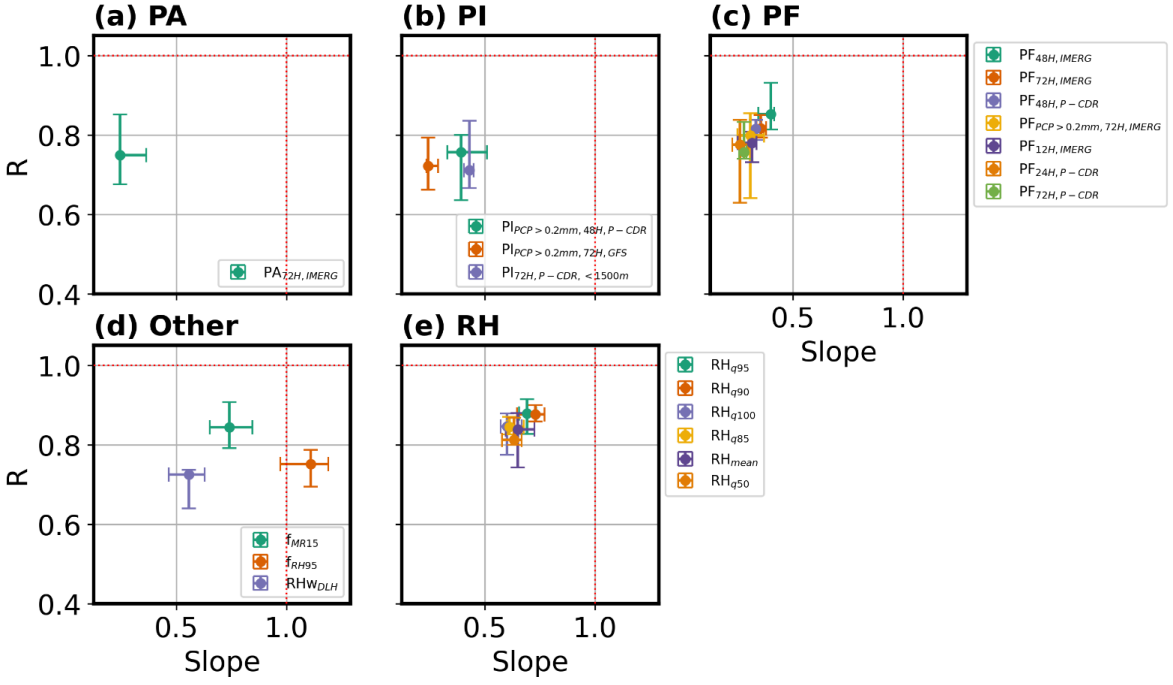


675

**Figure 2: Slope and Pearson correlation (R) values derived from linear regressions of observed (x) and predicted (y) ΔBC/ΔCO with error bars representing the 25th and 75th percentile values derived from k-fold cross validation (k=10) using stratified random sampling (Sect. 2.5). Ideal values are denoted by the red dashed lines such that a better predictor would fall closer to the intersection of the two lines. Only predictors with median R > 0.70 are shown. Note that PERSIANN-CDR has been abbreviated to P-CDR (b-c). Panels share the same X- and Y-axis limits.**

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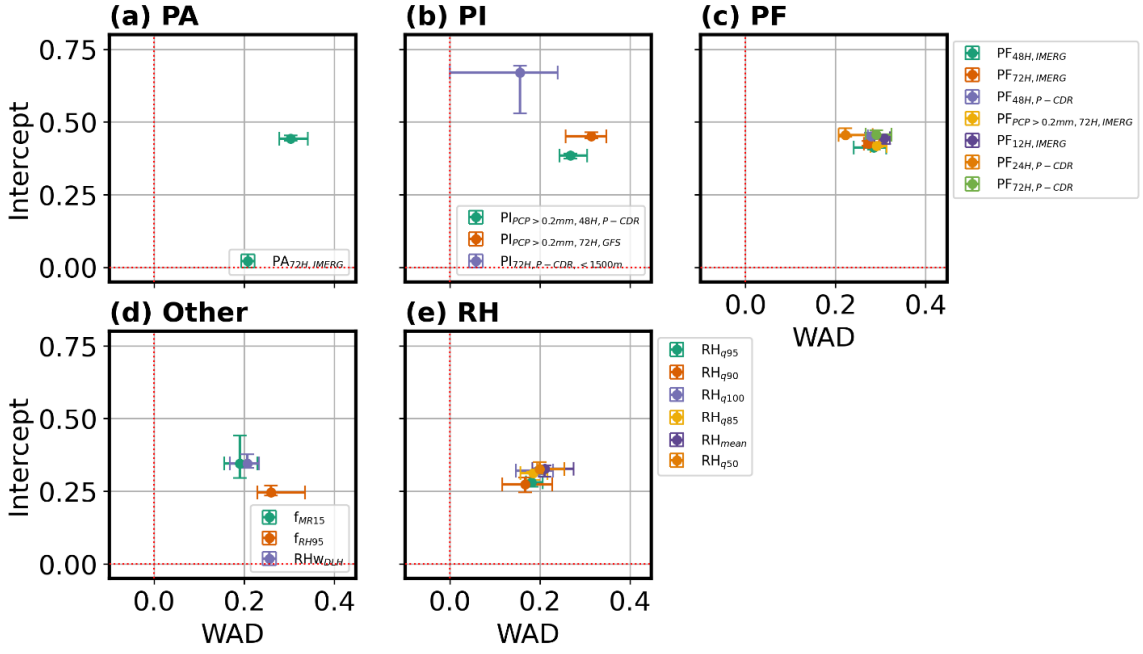


**Figure 3: Same as Fig. 2 but comparing intercept and weighted area difference (WAD, Sect. 2.5).**

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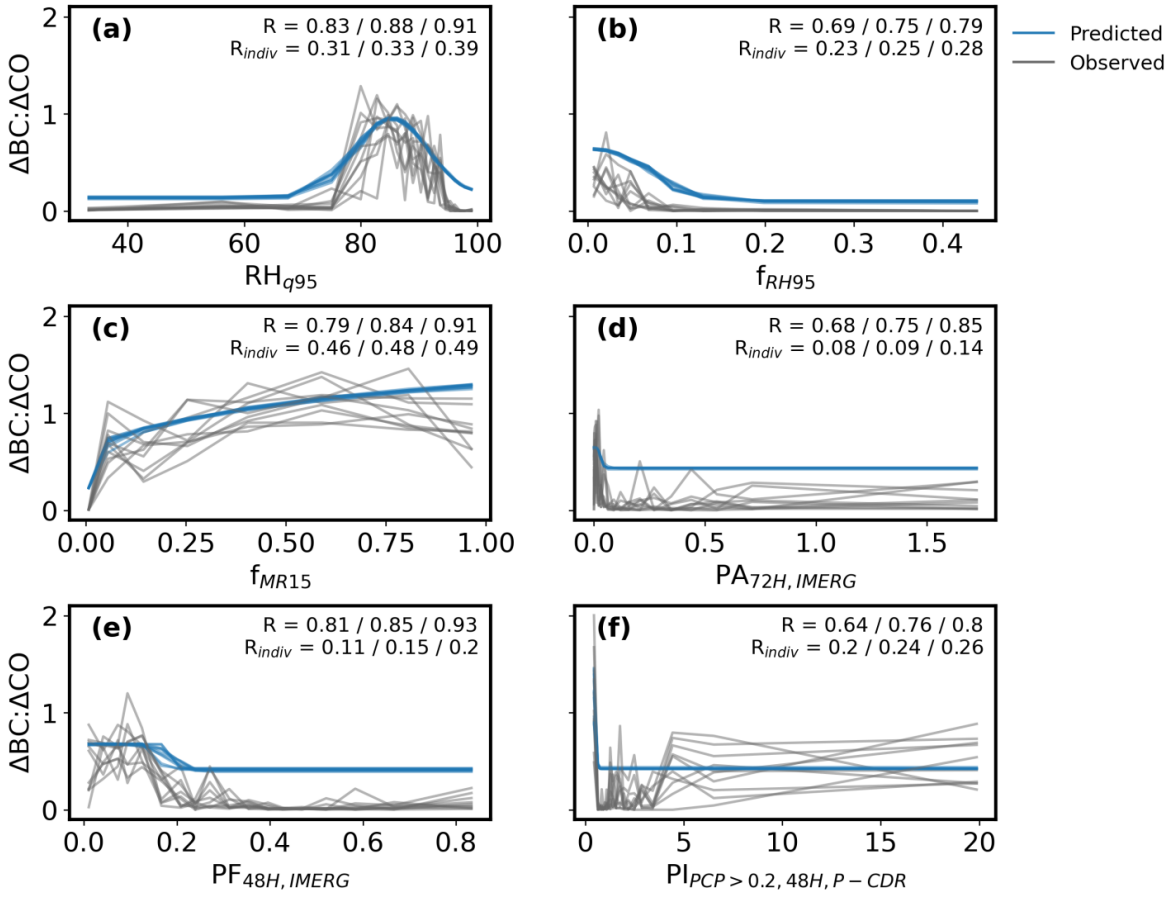


**Figure 4: Median values of observed (black) and predicted ΔBC/ΔCO (red) as a function of selected predictors for 10 iterations during k-fold cross-validation. Pearson correlations are annotated as 25th/50th/75th percentiles from k-fold iterations (k=10) calculated in two ways: comparing predicted and observed median ΔBC/ΔCO per 5th percentile block of the predictor (R) and comparing predicted and observed ΔBC/ΔCO for individual points ($R_{indiv}$). Panels share the same Y-axis limits.**

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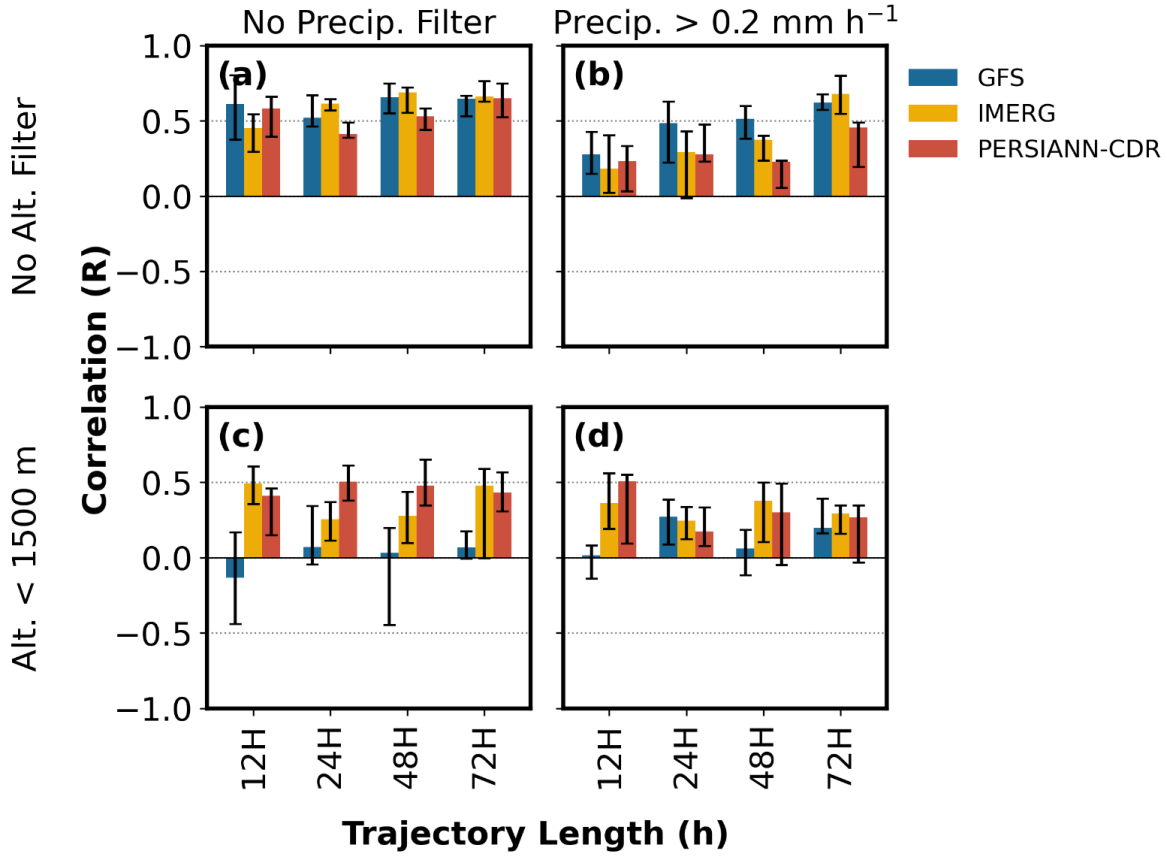


**Figure 5: Pearson correlations (R) between observed ΔBC/ΔCO and ΔBC/ΔCO predicted by accumulated precipitation along trajectories (APT) for different trajectory lengths and precipitation data products. Each panel refers to a combination of altitude and precipitation intensity filters. Panels share the same X- and Y-axis limits.**