

This study presents a valuable and comprehensive investigation into the atmospheric and terrestrial pathways, retention, and export of trifluoroacetate (TFA) at the catchment scale. The two-year dataset and multi-compartment sampling approach (precipitation, streamflow, springs, WWTPs) provide an important basis for understanding TFA dynamics. The manuscript is generally well written and organized; however, several methodological clarifications and interpretations should be addressed before publication.

Line 160–165:

Please clarify the rationale for choosing this separation column for TFA analysis. What are its advantages compared with other commonly used columns for PFAS analysis, such as the Hypersil Gold C18 column?

Line 165–170:

The role of 50 mM ammonium hydrogen carbonate in pure water as mobile phase A should be clarified. If the separation column is hydrophilic, the use of methanol as mobile phase B may not be ideal. Please confirm the column chemistry and justify the chosen mobile phase composition.

Line 175–176:

You mention Mill-Q blank samples as procedural blanks. Were these blanks also used to assess potential TFA contamination from the LC–MS system (e.g., tubing, fittings, internal components)? Please describe any specific cleaning or pre-conditioning procedures used to minimize TFA background signals from the instrument.

Line 190–195:

It is mentioned that the same separation and guard columns used for LC–MS analysis were also used for IC analysis of major anions and cations. Does this imply that the IC system could potentially be used for TFA determination as well? If so, was this tested or verified?

Additionally, please clarify whether “supplier” refers to the supplier of the IC instrument or of the columns. Did you determine quantification limits for each ion using your own calibration curves under actual operating conditions, which might be more accurate than supplier-provided values?

Line 275–276:

Please elaborate on the factors leading to the highest TFA levels in the Dreisam River during the 2023–2024 winter.

Figure 2:

Does the gray shading in panels (a–c) represent dry and wet conditions? Please specify this in the figure caption.

Line 303–304:

Based on the correlation data in Table 2, it seems that TFA showed positive correlations with all tracers except nitrate. Please confirm and revise accordingly.

Line 306:

The sentence “The same was true for the negative correlation with deuterium excess” is ambiguous. Please rephrase for clarity (e.g., “Similarly, TFA exhibited a negative correlation with deuterium excess”).

Line 364–365:

You state that “Potassium negatively correlated with TFA; however, concentrations were below LOQ and could not be reliably interpreted.” How was the correlation established if potassium concentrations were not quantifiable? Please clarify or reconsider this statement.

Line 366–367:

Given that the spring pH is around 6, the previously cited finding that “TFA sorption to soils decreased with increasing pH up to pH 5” (Richey et al., 1997) may not adequately explain the observed behavior at the Zipfeldobel spring. Please discuss this limitation.

Line 428–430:

Considering TFA’s high mobility, the lack of significant retention in water bodies contrasts with reported TFA retention in plants and soils (Likens et al., 1997; Berger et al., 1997). Please elaborate on possible mechanisms or environmental conditions explaining this discrepancy.

Line 430:

The sentence “Potentially, differences originate from the study design of both field experiments” should include more detail on what specific design differences (e.g., sampling frequency, soil types, hydrological setting) might explain the divergent results.

Line 447–449:

Please provide references supporting the statement that patterns/concentrations “were attributed to the distribution of TFA precursor molecules in the atmosphere.”

Line 380 vs. Line 470:

You stated that “our hypothesis of a temporal TFA storage, most likely associated with organic soil, seems valid,” yet later conclude that “We identified the organic soil zone as a primary TFA storage.” Since the data suggest only temporary accumulation, the latter conclusion may overstate the findings. Please rephrase to maintain consistency and avoid overinterpretation.

Line 475:

You compare TFA loads from farming activities with values reported for precursor PPP degradation in Joerss et al. (2024). If those values were derived from a different catchment, the comparison may not be meaningful. Please clarify whether the data are directly comparable.