Response to comments from the Referee 1:

I agree with the other reviewer of the manuscript, Bettina Schaefli (further refereed as BS), that it is too condensed in its current form. This factor makes the development of the system of equations' solution difficult to follow, and this must be improved. In addition, I am a bit concerned that this method and the presented illustration example have been reported previously by (Correa et al., 2019). Therefore, I strongly encourage the authors to: 1) mention openly whether their uncertainty estimation method is different than/similar to (the same?) the one applied in the aforementioned paper and ii) to apply their method to 1-2 additional illustration examples from the published literature or, at the very least, include the evaluation of outliers suggested by BS to differentiate this work from the one of (Correa et al., 2019).

R: We are very thankful to the Referee's useful remarks, which greatly helped to improve our Technical Note. We appreciate the comment that the Technical Note is condensed, and it should be extended and clarified to provide the community with an easy-to-follow reading material, mainly in the description of the system of equations', their development and solution.

Regarding 1) We thank the Referee for highlighting that we based our application on the example published in Correa et al (2019). However, the authors calculated the uncertainties based only on the application of a final equation. The main objective of this Technical Note is therefore to explicitly describe the mathematical development in all detail that allows the calculation of partial derivatives, degrees of freedom and confidence interval limits for each source fraction contribution as well as to provide the code and example data for their calculation and reproducibility.

Regarding 2) An evaluation of outliers as well as four additional examples from the same data set were included in the new version of the Technical Note. Additionally, a new figure (Figure. 2) was included showing Boxplots of end-members projected in the three-dimensional mixing space as a basis for clarity and a better understanding of the example calculations.

Please find at the end of this document the description of the examples and tables with input information (Table 1 to Table 4) and results (Table 5 to Table 8) that have been included and discussed in the Technical Note.

Major Comments:

The authors claim the robustness of their method (P.2, L.34), but do not discuss this consideration in comparison for example, to the commonly applied Gaussian error propagation approach. I think it would be great to evaluate and discuss this in the manuscript to add value to the usefulness of the presented methodology. This could also help differentiate this work from the work of (Correa et al., 2019).

R: We agree with the Reviewer that an exhaustive comparison of different methods should be attempted at some point, but after careful consideration we did not follow this suggestion here due to the length of this technical note. We on purpose used this format and not a full research paper to present our novel methodology in more mathematical detail than usual, step-by-step with an example application and we also provide the codes. This uncertainty assessment method was not presented in Correa et al. (2019), only parts of the dataset.

System of equations and resolution: please make sure to define clearly all of the notation in the set of equations to facilitate the readability of their resolution throughout the paper. All the considerations within the resolution of the system must also be clearly stated.

R: We appreciate this suggestion and have extended and updated the manuscript to clarify the notation and also to include more details in the descriptions of equations and variables to improve the readability of the technical note.

Specific comments:
Page 1, Lines 12-13: this sentence is incomplete. Please correct.
R: We have edited the phrase. It now reads: "[…], Bayesian approaches to estimate such source uncertainty only exist only sound methods for two and three sources."

P.1, L15: "dataset".

R: In this context the word "set" refers to the set of equations used to calculate the uncertainty of the source's contributions to a mixture, not to the data set, therefore we have omitted this change.

P.2, L.7: Replace "novel" by "the availability of".

R: This has been corrected to "the availability of"

P.2, L.12: Delete "novel".

R: The word "novel" has been deleted.

P.2, footnote: I think "M" refers to the mixture, not to a source. Correct if necessary.

R: This has been corrected to "mixture"

P.7, L.4: n is approximately 30 for each source, or among all sources

R: We have edited the phrase to clarify this point. It now reads: "[…] and spring water (SW) (n ~ 30, for each end-member) were collected".

P.7, L.21 and L.23: I think you refer to streamflow (or mixture, M), and not to spring water (SW). Correct if necessary.

R: We have edited the phrase to correct this error. "SW" was replaced by "M".

Table 2: report λ values
R: By λ we assumed that the Referee refers to degrees of freedom (γ). These values are reported in the Table 5.

Application examples

We have generated 6 examples to analyze the sensitivity of the uncertainty calculation to the source sample size, the artificial inclusion of outliers (upper and lower extremes) and the increased standard deviations of the source datasets. The first example considers 50% of the samples from each source. The median, standard deviation and sample size are input data (Table 2) to calculate the uncertainty ranges (Table 6). The second considers the remaining 50% of samples and was similarly executed (Table 2). In the third example, outliers were artificially included at the upper positive end of data sets for each source at each coordinate, respectively. The outliers consisted of twice the maximum positive value of the observed data (Table 3). Using the same criteria, the negative extremes were included in the fourth example (Table 3). - Sources affected by dispersed data clouds were taken into account by an increase in the standard deviation. We considered two cases, the first, in the example five, increasing three times the value of the standard deviation of the initial data set (Table 4) and finally, increasing the standard deviation five times for the sixth example (Table 4).

Table 1. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional space for the study period 2013–2014.

End-member		Coordinates			Naming
		U1	U2	U3	in equations
SW (n = 25)	median	26,25	7,29	7,00	A
	std.dev.	0,46	0,36	0,39	
HS (n = 33)	median	0,23	5,48	1,97	B
	std.dev.	0,85	1,29	0,69	
AN (n = 37)	median	-2,24	-3,93	3,71	C
	std.dev.	0,55	0,58	0,45	
RF (n = 36)	median	-5,38	-6,10	-4,84	D
	std.dev.	0,27	0,56	0,15	
Stream (n = 257)	median	-0,61	-1,04	0,94	M
	std.dev.	2,06	1,10	0,66	

Table 2. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional considering 50% of the data sets (examples 1 and 2)

Naming in equations		1)	End member	Coordinates U1	U2	U3	2)	End member	Coordinates U1	U2	U3
A	median		SW	26.18	7.29	6.66		SW	26.28	7.29	7.1
	std.dev.		(n = 12)	0.34	0.39	0.48		(n = 13)	0.51	0.36	0.21
B	median		HS	0.23	5.41	1.87		HS	0.28	5.9	2.26
	std.dev.		(n = 17)	0.74	1.19	0.52		(n = 17)	0.96	1.33	0.74
C	median		AN	-2.37	-3.93	3.69		AN	-2.2	-3.94	3.89
	std.dev.		(n = 19)	0.59	0.4	0.49		(n = 19)	0.46	0.73	0.41
D	median		RF	-5.37	-6.26	-4.78		RF	-5.35	-5.99	-5.01
	std.dev.		(n = 18)	0.26	0.58	0.07		(n = 18)	0.28	0.53	0.15
M	median		Stream	-0,61	-1,04	0,94		Stream	-0,61	-1,04	0,94
	std.dev.		(n = 257)	2,06	1,10	0,66		(n = 257)	2,06	1,10	0,66

The example 1) considers the initial 50% and 2) the remaining 50% of the sample sets.

Table 3. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional including artificial outliers (examples 3 and 4)

Naming in equations		3)	End member	Coordinates U1	U2	U3	4)	End member	Coordinates U1	U2	U3
A	median		SW	26.25	7.3	7.02		SW	26.21	7.29	6.95
	std.dev.		(n = 26)	5.51	1.73	1.68		(n = 26)	10.28	2.87	2.54
B	median		HS	0.27	5.47	1.98		HS	0.23	5.45	1.97
	std.dev.		(n = 34)	0.99	2.45	1.03		(n = 34)	1.12	1.99	0.8
C	median		AN	-2.24	-3.92	3.79		AN	-2.26	-3.95	3.74
	std.dev.		(n = 38)	0.78	1.17	0.92		(n = 38)	1.07	1.43	1.15
D	median		RF	-5.36	-6.08	-4.84		RF	-5.37	-6.11	-4.86
	std.dev.		(n = 37)	1.7	1.89	1.58		(n = 37)	1.09	1.42	0.94
M	median		Stream	-0,61	-1,04	0,94		Stream	-0,61	-1,04	0,94
	std.dev.		(n = 257)	2,06	1,10	0,66		(n = 257)	2,06	1,10	0,66

The example 3) considers outliers included at the positive extreme of the dataset of each source and 4) outliers included at the negative extreme.

Table 4. Median and enlarged standard deviation (std.dev.) of end-members and stream projected in three-dimensional (examples 5 and 6)

Naming in equations		5)	End member	Coordinates U1	U2	U3	6)	End member	Coordinates U1	U2	U3
A	median		SW	26,25	7,29	7,00		SW	26,25	7,29	7,00
	std.dev.		(n = 25)	1.39	1.07	1.19		(n = 25)	2.32	1.78	1.99
B	median		HS	0,23	5,48	1,97		HS	0,23	5,48	1,97
	std.dev.		(n = 33)	2.56	3.87	2.06		(n = 33)	4.27	6.45	3.43
C	median		AN	-2,24	-3,93	3,71		AN	-2,24	-3,93	3,71
	std.dev.		(n = 37)	1.65	1.73	1.34		(n = 37)	2.75	2.88	2.24
D	median		RF	-5,38	-6,10	-4,84		RF	-5,38	-6,10	-4,84
	std.dev.		(n = 36)	0.8	1.69	0.46		(n = 36)	1.34	2.81	0.77
M	median		Stream	-0,61	-1,04	0,94		Stream	-0,61	-1,04	0,94
	std.dev.		(n = 257)	2,06	1,10	0,66		(n = 257)	2,06	1,10	0,66

The example 5) considers 3-times the standard deviation of the original data set and 6) 5-times the standard deviation of the original data set.

Table 5. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom calculated for the study period 2013–2014

	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution	0.06	0.3	0.35	0.29
Upper 95% confidence limit	0.21	0.57	0.58	0.46
Lower 95% confidence limit	0.00	0.03	0.12	0.12
Degrees of freedom	291	536	749	628

Table 6. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed considering 50% of the data sets

	1)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	2)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.28		0.06	0.28	0.35	0.3
Upper 95% confidence limit		0.21	0.57	0.58	0.45		0.21	0.55	0.58	0.46
Lower 95% confidence limit		0.00	0.03	0.12	0.11		0.00	0.02	0.12	0.14
Degrees of freedom		289	493	676	589		288	491	679	537

The example 1) was computed considering the initial 50% and 2) the remaining 50% of the sample sets.

Table 7. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed after including artificial outliers

	3)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	4)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.29		0.06	0.3	0.35	0.29
Upper 95% confidence limit		0.22	0.62	0.64	0.5		0.22	0.61	0.63	0.49
Lower 95% confidence limit		0.00	0.00	0.06	0.08		0.00	0.00	0.07	0.08
Degrees of freedom		350	448	640	529		353	554	757	621

The example 3) was computed after including outliers at the positive extreme of the dataset and 4) including outliers at the negative extreme.

Table 8. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed with enlarged standard deviations

	5)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	6)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.29		0.06	0.3	0.35	0.29
Upper 95% confidence limit		0.23	0.68	0.69	0.52		0.26	0.83	0.83	0.61
Lower 95% confidence limit		0.00	0.00	0.01	0.05		0.00	0.00	0.00	0.00
Degrees of freedom		372	225	362	312		335	122	211	172

The example 5) was computed considering 3-times the standard deviation of the original data set and 6) 5-times the standard deviation of the original data set.

Response to comments from the Referee 2:

Major comments:
The main problem with this technical note is the lack of definitions and explanations of variables and notations. Albeit I understand that a technical note is supposed to be short, if the authors want this work to be utilized by other geoscientists and hydrologists, they need to improve the readability of the technical note.

R: We appreciate the Reviewer's comment in line with another Reviewer’s evaluation and have followed his/her suggestions throughout the document. We now included more details on descriptions of equations, variables and notations to improve the readability of the technical note.

If I understand correctly, this technical note details the methodology used by Correa et al. (2019). It would be great if the authors could be more upfront about this in the technical note.

R: We are grateful for the comment and we agree, in the new version of the Technical Note it is clearly specified that the methodology developed here is the one applied in Correa et al. (2019b).
It now reads: “We illustrate this application on the study case published in Correa et al. (2019b), where the authors presented the uncertainty analysis of sources contributions results […]”.

The Matlab script is relatively hard to follow and understand, due to insufficient comments and documentation:
Naming of Matlab Files: Matlab filenames starting with a number cannot be run by Matlab. Please rename in order to avoid confusion for unexperienced Matlab users.
Name of csv file is different in repository than as used in matlab script (5_data.csv rather than data.csv). Please make consistent.
The Matlab Script is not very well documented.
Neither the script directly, nor the readme pdf explain the actual inputs (only some cryptic A/B/C/D/M without explanations; one has to refer to Table 1 to understand the setup of the file) nor the outputs.
Consequently, the script cannot be used without reading the manuscript in detail.
It would furthermore be helpful to reference the code lines to the equations of the manuscript and state in the script what each command is doing.
Please also reference the manuscript in the readme.pdf and the main script.
Specific Comments:

R: We are very grateful that the reviewer highlights the lack of readability in the MatLab script and its documentation. We intended to use the script along with the technical note where the definitions of terms are stated, however, we have included a detailed description in the documentation of the script. Additionally, we have maintained consistency between the repository and the code.
The number of equations in the manuscript have been included in the code (method.m) for reference, along with a description of what each equation does.
The manuscript (under review) will be referenced in the readme file.
Please find at the end of this document, the updated readme and method files.

Specific Comments:
p.1 line 13: the grammar of this sentence is wrong/the sentence is incomplete.

R: We have edited the phrase. It now reads: “[…], Bayesian approaches to estimate such source uncertainty only exist only sound methods for two and three sources.”

p.1 lines 15-17: There is no connection between the two sentences, in spite of the “however”. Sentence 1 talks about large tracer sets from four water sources (but says nothing about the number of tracers), Sentence 2 says the approach can be generalized

to any number of tracers. Please make this clearer.

R: We have updated this section as suggested: "We illustrate the method to compute individual uncertainties in the calculation of source contributions to a mixture, particularly with an example from hydrology, where a 14-tracer set from water sources and streamflow from a tropical, high-elevation catchment were used. Moreover, this method has the potential to be generalized to any number of tracers in a wide range of disciplines."

p.5 line 1-2: the reference of Taylor, 1982 should probably follow directly after "Taylor series approximation".

R: We have edited the phrase. It now reads: "[…], the first-order Taylor series approximation (Taylor, 1982) for the variance […]."

p.7 line 2: the specification of n=270 is no useful without also specifying the number of different streams sampled.

R: We thank the reviewer for pointing out a typographical error, the correct number of samples is 257 and the number of streams sampled is 5, the phrase was updated: "Streamwater samples from 5 nested streams were collected weekly from March 2013 to April 2014 (n=257) and at a higher frequency during experimental campaigns."

References: Correa et al, 2018, SciTotEnv should actually be Correa et al. 2019.

R: We updated the reference: Correa, A., Breuer, L., Crespo, P., Célleri, R., Feyen, J., Birkel, C., Silva, C. and Windhorst, D.: Spatially distributed hydro-chemical data with temporally high-resolution is needed to adequately assess the hydrological functioning of headwater catchments, Science of The Total Environment, 651, 1613–1626, doi:10.1016/j.scitotenv.2018.09.189, 2019b.

Table 1: Footnote: "three- axes". I believe this should be three-dimensional space.
R: We have edited the footnote. It now reads: "Coordinates of end-members and stream (mixture) medians in three-dimensional space (U1, U2 and U3). n represents the sample size"

Table 2: Some of the values provided here do not match up with those calculated by the MATLAB Code. Please verify.

R: We have verified the result from the MatLab code, and the values presented in table 2 are correct. However, in the Zenodo platform, the data.csv preview shows the data rounded to a decimal and a missing column (Stvd U2), it is necessary to download the.csv file to get the complete data.
Updating the code, we will improve the visualization of the data.

Readme file

These codes estimate the uncertainty of individual end-member (source) contributions to streams (mixture) based on a multi-tracer set in a three-dimensional space.
The method.m code shows step-by-step calculations of partial derivatives, degrees of freedom, t-Student and confidence interval limits for each source fraction.
method.m uses the functions Yx.m and dYzdyx.m for its execution.
A, B, C and D represent the set of sources and M the mixture.
Please refer to Correa et al., (2019) in Correa, A., Ochoa-Tocachi, D. and Birkel, C.: Technical note:
Uncertainty in multi-source partitioning using large tracer data sets, Hydrology and Earth System Sciences Discussions, 1–14, doi:https://doi.org/10.5194/hess-2019-197, 2019 for a very detailed description of the used notation, equations and variables for this example.
The equation numbers used in the code method.m refer to the corresponding ones in Correa et al., (2019).

Instructions:
Enter data for the median of end-members and mixture, their standard deviations and sample size, all projected in the three-dimensional PCA-space (U-space in the referred Technical Note). The file must be named data.csv and follows an order similar to the one presented in this example:

	Median in U1	Median in U2	Median in U3	Stvd in U1	Stvd in U2	Stvd in U3	No. samples	No. samples	No. samples
A	26.25	7.29	7.00	0.46	0.36	0.4	25	25	25
B	0.23	5.47	1.98	0.85	1.29	0.69	33	33	33
C	-2.24	-3.93	3.74	0.55	0.58	0.45	37	37	37
D	-5.38	-6.1	-4.85	0.27	0.56	0.15	36	36	36
M	-0.61	-1.04	0.94	2.07	1.1	0.66	257	257	257

Please do not include rows and column names in the data.csv file, here is used for merely visual purposes of the example.

Run the method.m code.
It is recommended to keep all files in a common directory (method.m; dYzdyx.m; Yx.m and data.csv)
After executing the code, the fractions of the contribution of each end member to the mixture are calculated, as well as the
degrees of freedom and upper and lower limits of uncertainties associated with their contribution.

Note: the order of the inputs (end members) is reflected in the outputs (fractions of contribution and uncertainties).

method file

```
clear, clc;
format short;
%% Data
%% DATA reads the data.csv file with the input information
DATA = csvread('data.csv');
% Ms
Z = [DATA(:,1:3), ones(5,1)];
Z = transpose(Z);
dataM = Z(:,end);
Z = Z(:, 1:end-1);
% Variance
vZ = transpose (DATA(:,4:6)).^(2);
% Sample QTY
nZ = transpose (DATA(:,7:9));
%% Compute the fractions of sources (A, B, C, D) contribution to the mixture (M) (Eq. 2 and implicitly from Eq. 3)
% x in {A,B,C,D,M} = {1,2,3,4,5}
% y in {delta,lambda,phi} = {1,2,3}
f = zeros(4,1);
DYx = @(y,x) Yx(y,x, Z, dataM);
Num = (DYx(1,5)-DYx(2,5))*(DYx(3,3)-DYx(1,3)) - (DYx(1,3)-DYx(2,3))*(DYx(3,5)-DYx(1,5));
Den = (DYx(1,1)-DYx(2,1))*(DYx(3,3)-DYx(1,3)) - (DYx(1,3)-DYx(2,3))*(DYx(3,1)-DYx(1,1));
f(1) =  Num/Den;
f(3) = ((DYx(1,5)-DYx(2,5))-(DYx(1,1)-DYx(2,1))*f(1))/(DYx(1,3)-DYx(2,3));
f(2) = DYx(1,5) - (DYx(1,3)*f(3) + DYx(1,1)*f(1));
f(4) = 1 - (f(1) + f(2) + f(3));
%% Compute the partial derivatives for fA, fC, fB and fD (Eq. 4 and implicitly from Eq. 5 to Eq. 8)
% x in {A, B, C, D, M} = {1,2,3,4,5}
% Y,y in {delta,lambda,phi} = {1,2,3}
%% dfAdyx, presents the partial derivative for fA (Eq. 9)
DdYzdyx = @(Y,z,y,x) dYzdyx(Y,z,y,x,Z,dataM);
dfAdyx = @(y,x) Den^(-2)*( ...
   ((DYx(2,3)-DYx(1,3))*(DdYzdyx(3,5,y,x) - DdYzdyx(1,5,y,x)) + ...
    (DYx(3,5)-DYx(1,5))*(DdYzdyx(2,3,y,x)-DdYzdyx(1,3,y,x)) - ...
    (DYx(3,3)-DYx(1,3))*(DdYzdyx(2,5,y,x)-DdYzdyx(1,5,y,x)) - ...
    (DYx(2,5)-DYx(1,5))*(DdYzdyx(3,3,y,x)-DdYzdyx(1,3,y,x)))*Den - ...
    ((DYx(2,3)-DYx(1,3))*(DdYzdyx(3,1,y,x)-DdYzdyx(1,1,y,x)) + ...
    (DYx(3,1)-DYx(1,1))*(DdYzdyx(2,3,y,x)-DdYzdyx(1,3,y,x)) - ...
    (DYx(3,3)-DYx(1,3))*(DdYzdyx(2,1,y,x)-DdYzdyx(1,1,y,x)) - ...
    (DYx(2,1)-DYx(1,1))*(DdYzdyx(3,3,y,x)-DdYzdyx(1,3,y,x)))*Num);
 dfCdyx = @(y,x) ((DYx(1,3)-DYx(2,3))^(-2))*( ...
   ((DdYzdyx(1,5,y,x)-DdYzdyx(2,5,y,x)) - ...
   (DdYzdyx(1,1,y,x)-DdYzdyx(2,1,y,x))*f(1) - ...
   (DYx(1,1)-DYx(2,1))*dfAdyx(y,x))*(DYx(1,3)-DYx(2,3)) - ...
   (DdYzdyx(1,3,y,x)-DdYzdyx(2,3,y,x))*( ...
   (DYx(1,5)-DYx(2,5))-(DYx(1,1)-DYx(2,1))*f(1)));
dfBdyx = @(y,x) DdYzdyx(1,5,y,x) - DdYzdyx(1,3,y,x)*f(3) - ...
   DYx(1,3)*dfCdyx(y,x) - DdYzdyx(1,1,y,x)*f(1) - DYx(1,1)*dfAdyx(y,x);
dfDdyx = @(y,x) -dfCdyx(y,x)-dfBdyx(y,x)-dfAdyx(y,x);
%% Compute the variancefor each end-member fraction, fA, fB, fC and fD respectively (Eq. 10 and Eq. 13)
% x in {A,B,C,D,M} = {1,2,3,4,5}
% y in {delta,lambda,phi} = {1,2,3}
v = zeros(4,1);
```

```
for x = 1:5
   for y = 1:3
      v(1) = v(1) + (dfAdyx(y,x)^2)*vZ(y,x);
      v(2) = v(2) + (dfBdyx(y,x)^2)*vZ(y,x);
      v(3) = v(3) + (dfCdyx(y,x)^2)*vZ(y,x);
      v(4) = v(4) + (dfDdyx(y,x)^2)*vZ(y,x);
   end
end
%% Satterthwaite degrees of freedom for each end-member fraction (Eq. 12 and Eq. 14).
% x in {A,B,C,D,M} = {1,2,3,4,5}
% y in {delta,lambda,phi} = {1,2,3}
g = zeros(4,1);
for x = 1:5
   for y = 1:3
       g(1) = g(1) + (((dfAdyx(y,x)^2)*vZ(y,x))^2)/(nZ(y,x)-1);
       g(2) = g(2) + (((dfBdyx(y,x)^2)*vZ(y,x))^2)/(nZ(y,x)-1);
       g(3) = g(3) + (((dfCdyx(y,x)^2)*vZ(y,x))^2)/(nZ(y,x)-1);
       g(4) = g(4) + (((dfDdyx(y,x)^2)*vZ(y,x))^2)/(nZ(y,x)-1);
   end
end
g = (v.^2)./g;
%% Student's t value(two-tailed) to compute 95% confidence intervals (Walpole et al., 2017)
t = tinv(0.95,g);
%% Compute the upper and lower confidence interval limits for each end-member fraction (Eq. 15)
ulim = min(1,f + t.*sqrt(v));
llim = max(0,f - t.*sqrt(v));
%% Present results:
%% f, fractions of sources (A,B,C,D) contribution to the mixture (M),
%% g, degrees of freedom for each end-member fraction
%% ulim and llim, upper and lower confidence interval limits for each end-member fraction
f
g
ulim
llim
```

List of all relevant changes

- The abstract has been improved
- The introductory section was expanded following the referees' recommendations, we explicitly explained how the uncertainty estimation method relates to Correa et al 2019 (line 55).
- The revised manuscript has been expanded to clarify the notation and includes more details in the description of equations and variables, all to improve the comprehensibility of the Technical Note.
- In the methods section, we have added the equation to calculate the upper and lower limits of the uncertainty band (line 91).
- A new figure (Figure 2) was included to show via Boxplots, the initial set of end members projected into the three-dimensional mixing space as a basis for a clear understanding of the new application examples (line 135).
- We have generated 6 examples to assess the sensitivity of the uncertainty calculation to the source sample size, the artificial inclusion of outliers (upper and lower extremes) and the increased standard deviations of the source datasets. The examples were presented in Tables 2 to 4 and 6 to 8, analysed and discussed (line 320 – 355).
- The methodology was presented as an alternative to the simple sum of analytical errors or the Bayesian approach. We are confident that this Technical Note provides a useful tool (methodology plus MatLab code) to help the community to better conceptualize the source contributions to the mixture within a robust uncertainty framework.
- The specific corrections suggested by the Referees were fully covered.

Technical note: Uncertainty in multi-source partitioning using large tracer data sets

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Abstract

The availability of large tracer data sets opened up the opportunity to investigate multiple source contributions to a mixture. However, the source contributions may be uncertain and apart from Bayesian approaches to estimate such source uncertainty only exist ~~only~~ sound methods for two and three sources. We expand these methods developing an uncertainty estimation method for four sources based on multiple tracers as input data. Taylor series approximation is used to solve the set of linear mass balance equations. We illustrate the method to compute individual uncertainties in the calculation of source contributions to a mixture, particularly with an example from hydrology, where a 14-tracer set from water sources and streamflow from a tropical, high-elevation catchment were used. Moreover, this method has the potential to be generalized to any number of tracers across a range of disciplines.~~We illustrate the method with an example from hydrology, where we use a large tracer set from four water sources contributing to streamflow in a tropical, high-elevation catchment. However, our uncertainty estimation method can be generalized to any number of tracers across a range of disciplines.~~

1. Introduction

Tracer applications have dramatically increased over recent years across a wide range of disciplines (West et al., 2010). Applications in hydrology (Hooper, 2003; James and Roulet, 2006; Kirchner and Neal, 2013), ecology (Phillips and Gregg, 2003; Semmens et al., 2009b), anthropology (Ehleringer et al., 2008), conservation biology (Bicknell et al., 2014), nutrition (Magaña-Gallegos et al., 2018), environmental and ecosystem science (Bartov et al., 2013; Granek et al., 2009), and erosion and sediment transportation (Davies et al., 2018) have been the most prominent. Such a widespread use of tracers was mainly facilitated by the availability of~~novel~~ analytical techniques that provide high sensitive, rapid multi-element analysis at lower cost (Falkner et al., 1995). For example, the use of inductively coupled plasma mass spectrometry (ICP-MS) as one of the leading analytical techniques for elemental analysis (Helaluddin et al., 2016), led to the availability and use of large tracers sets (elements) in hydrological studies (Barthold et al., 2017; Belli et al., 2017; Correa et al., 2017; Kirchner and Neal, 2013; Mimba et al., 2017)~~(Barthold et al., 2017; Belli et al., 2017; Correa et al., 2018; Kirchner and Neal, 2013; Mimba et al., 2017)~~. Trace elements together with water stable isotopes (~~novel~~ Cavity Ringdown Laser Absorption Spectroscopy paved the way: (Berman et al., 2009; Lis et al., 2008)) as well as physical-chemical water parameters (e.g. electrical conductivity and pH) are now often used to improve understanding of hydro-geochemical cycles, flow pathways and runoff generation in hydrology. Furthermore, mixing models based on mass balance equations are widely-applied to identify the dominant sources and their dynamics as components of a mixture.

In hydrological mixing models the composition of the stream is assumed to be an integrated mixture of signatures of different sources (Christophersen et al., 1990). The proportional contributions of n+1 sources to the stream can be uniquely determined using n different tracers (Christophersen & Hooper, 1992). Bayesian methods have been developed to identify multiple (> 3) sources and compute their contributions to a mixture in a two-dimensional space (Parnell et al., 2010; Stock et al., 2018). In this case a unique solution is not feasible and a higher uncertainty is attributed to the model (Phillips and Gregg, 2001, 2003). On the other hand, End Member Mixing Analysis (EMMA) (Hooper, 2003) was developed to use multiple tracers as input, and therefore, allows for a multi-dimensional space that potentially increases the number of identifiable sources (Barthold et al., 2011; Inamdar et al., 2013; Liu et al., 2004). Additionally, the use of multiple tracers can avoid bias and subjectivity in the input information. Therefore, EMMA provides a robust and complete conceptualization of catchment functioning and source interactions during runoff generation (Iwasaki et al., 2015). However, despite its benefits, the EMMA approach lacks a formal methodology to assess the uncertainty of multiple end-members (Delsman et al., 2013) and to assess individual uncertainties in the calculation of source contributions to a stream.

To our knowledge, the uncertainty estimation of source contributions to streams is based on Gaussian error propagation (Genereux, 1998) and was so far only calculated using one or two tracers simultaneously (MixSIAR: Parnell et al., 2010; Phillips & Gregg, 2001; Semmens, Moore, et al., 2009). Alternatively, when the number of sources is higher, the uncertainty is usually based on the sum of analytical errors, elevation effects and the spatial variability of end-member concentrations (Uhlenbrook and Hoeg, 2003). Hence, we propose a novel and robust methodology to estimate the uncertainty of individual end-member (source) contributions to streams (mixture) based on a multi-tracer set in a three-dimensional space defined by a Principal Component Analysis. ~~We outline and explain the step by step development of the mathematical procedure and give an example application including MatLab codes using a large multi-tracer data set from an experimental catchment in Ecuador.~~

We illustrate this application using data from Correa et al. (2019b), where the authors calculated the uncertainties only based on the application of a final equation without disclosing any details in the calculation and methodology used. The main objective of this Technical Note is therefore to explicitly describe the mathematical development in all detail that allows the calculation of partial derivatives, degrees of freedom and confidence interval limits for each source fraction contribution as well as to provide the code and example data for their calculation and reproducibility.

2. Uncertainty estimation method development

In this section, the uncertainty estimation method presented in Phillips and Gregg, (2001) is expanded for four source contributions to the mixture.

Let $\mathcal{C}$, represents the set of sources: A, B, C and D, and mixture M, $\mathcal{C} = \{A, B, C, D, M\}$. In the following equations, $x \in \mathcal{C}$, $y \in \{\overline{\delta}, \overline{\lambda}, \overline{\phi}\}$ and $z \in \{A, M, C\}$. x, y and z are variables that belong to the sets: x to the set of A, B, C, D and mixture M, y to the set of medians of every projected source and mixture in each principal component $\overline{\delta}, \overline{\lambda}, \overline{\phi}$ respectively of the used sub index and z to the set of A, M and C.

If the system is composed of Eq. (1)

$$\begin{cases} \overline{\delta}_A f_A + \overline{\delta}_B f_B + \overline{\delta}_C f_C + \overline{\delta}_D f_D = \overline{\delta}_M \\ \overline{\lambda}_A f_A + \overline{\lambda}_B f_B + \overline{\lambda}_C f_C + \overline{\lambda}_D f_D = \overline{\lambda}_M \\ \overline{\phi}_A f_A + \overline{\phi}_B f_B + \overline{\phi}_C f_C + \overline{\phi}_D f_D = \overline{\phi}_M \\ f_A + f_B + f_C + f_D = 1 \end{cases} \quad \text{Eq.(1)}$$

where f_A, f_B, f_C and f_D represent the contribution fraction of sources A, B, C and D respectively to the mixture M and Eq. (1) has solution[1] for $f_A, f_B, f_C, f_D > 0$, they take the following form:

$$\begin{aligned} f_A &= \frac{(\Phi_M - \Delta_M)(\Lambda_C - \Delta_C) - (\Lambda_M - \Delta_M)(\Phi_C - \Delta_C)}{(\Phi_A - \Delta_A)(\Lambda_C - \Delta_C) - (\Lambda_A - \Delta_A)(\Phi_C - \Delta_C)} = \frac{Num}{Den} \\ f_C &= \frac{(\Delta_M - \Lambda_M) - (\Delta_A - \Lambda_A) f_A}{(\Delta_C - \Lambda_C)} \\ f_B &= \Delta_M - (\Delta_C f_C + \Delta_A f_A) \\ f_D &= 1 - (f_C + f_B + f_A) \end{aligned} \quad \text{Eq.(2)}$$

where

$$\Delta_x = \frac{\overline{\delta}_x - \overline{\delta}_D}{\overline{\delta}_B - \overline{\delta}_D}, \ \Lambda_x = \frac{\overline{\lambda}_x - \overline{\lambda}_D}{\overline{\lambda}_B - \overline{\lambda}_D}, \ \Phi_x = \frac{\overline{\phi}_x - \overline{\phi}_D}{\overline{\phi}_B - \overline{\phi}_D}. \quad \text{Eq.(3)}$$

The partial derivatives of Eq. (2) are given by:

[1] The system has a solution if the vector of ~~source~~ mixture M is on the polyhedron generated by the vectors of sources A, B, C, D such that $\sum_x f_x = 1$.

$$
\begin{aligned}
\frac{\partial f_A}{\partial y_x} &= \frac{1}{Den^2}\Bigg[\Bigg[(\Lambda_C-\Delta_C)\left(\frac{\partial \Phi_M}{\partial y_x}-\frac{\partial \Delta_M}{\partial y_x}\right)+(\Phi_M-\Delta_M)\left(\frac{\partial \Lambda_C}{\partial y_x}-\frac{\partial \Delta_C}{\partial y_x}\right) \\
&\quad -(\Phi_C-\Delta_C)\left(\frac{\partial \Lambda_M}{\partial y_x}-\frac{\partial \Delta_M}{\partial y_x}\right)-(\Lambda_M-\Delta_M)\left(\frac{\partial \Phi_C}{\partial y_x}-\frac{\partial \Delta_C}{\partial y_x}\right)\Bigg] Den \\
&\quad -\Bigg[(\Lambda_C-\Delta_C)\left(\frac{\partial \Phi_A}{\partial y_x}-\frac{\partial \Delta_A}{\partial y_x}\right)+(\Phi_A-\Delta_A)\left(\frac{\partial \Lambda_C}{\partial y_x}-\frac{\partial \Delta_C}{\partial y_x}\right) \\
&\quad -(\Phi_C-\Delta_C)\left(\frac{\partial \Lambda_A}{\partial y_x}-\frac{\partial \Delta_A}{\partial y_x}\right)-(\Lambda_A-\Delta_A)\left(\frac{\partial \Phi_C}{\partial y_x}-\frac{\partial \Delta_C}{\partial y_x}\right)\Bigg] Num\Bigg] \\
\frac{\partial f_C}{\partial y_x} &= \frac{1}{(\Delta_C-\Lambda_C)^2}\Bigg[\left[\left(\frac{\partial \Delta_M}{\partial y_x}-\frac{\partial \Lambda_M}{\partial y_x}\right)-\left(\frac{\partial \Delta_A}{\partial y_x}-\frac{\partial \Lambda_A}{\partial y_x}\right)f_A-(\Delta_A-\Lambda_A)\frac{\partial f_A}{\partial y_x}\right](\Delta_C-\Lambda_C) \\
&\quad -\left(\frac{\partial \Delta_C}{\partial y_x}-\frac{\partial \Lambda_C}{\partial y_x}\right)[(\Delta_M-\Lambda_M)-(\Delta_A-\Lambda_A)f_A]\Bigg], \\
\frac{\partial f_B}{\partial y_x} &= \frac{\partial \Delta_M}{\partial y_x}-\frac{\partial \Delta_C}{\partial y_x}f_C-\Delta_C\frac{\partial f_C}{\partial y_x}-\frac{\partial \Delta_A}{\partial y_x}f_A-\Delta_A\frac{\partial f_A}{\partial y_x}, \\
\frac{\partial f_D}{\partial y_x} &= -\frac{\partial f_C}{\partial y_x}-\frac{\partial f_B}{\partial y_x}-\frac{\partial f_A}{\partial y_x}
\end{aligned}
\qquad \text{Eq.(4)}
$$

It is trivial that~~where~~

$$
\frac{\partial \Delta_z}{\partial w_x}=0,\ w\in\{\bar{\lambda},\bar{\phi}\};\ \frac{\partial \Lambda_z}{\partial w_x}=0,\ w\in\{\bar{\delta},\bar{\phi}\};\ \frac{\partial \Phi_z}{\partial w_x}=0,\ w\in\{\bar{\delta},\bar{\lambda}\}. \qquad \text{Eq.(5)}
$$

where

$$
\frac{\partial \Delta_z}{\partial \bar{\delta}_x} = \left(\bar{\delta}_B-\bar{\delta}_D\right)^{-1}\begin{cases}1 & z\in\{A,C,M\} \text{ and } x=z \\ -\Delta_z & z\neq B \text{ and } x=B \\ \Delta_z-1 & z\neq D \text{ and } x=D \\ 0 & otherwise\end{cases}, \qquad \text{Eq.(6)}
$$

$$
\frac{\partial \Lambda_z}{\partial \bar{\lambda}_x} = \left(\bar{\lambda}_B-\bar{\lambda}_D\right)^{-1}\begin{cases}1 & z\in\{A,C,M\} \text{ and } x=z \\ -\Lambda_z & z\neq B \text{ and } x=B \\ \Lambda_z-1 & z\neq D \text{ and } x=D \\ 0 & otherwise\end{cases} \quad \text{and} \qquad \text{Eq.(7)}
$$

$$
\frac{\partial \Phi_z}{\partial \bar{\phi}_x} = \left(\bar{\phi}_B-\bar{\phi}_D\right)^{-1}\begin{cases}1 & z\in\{A,C,M\} \text{ and } x=z \\ -\Phi_z & z\neq B \text{ and } x=B \\ \Phi_z-1 & z\neq D \text{ and } x=D \\ 0 & otherwise\end{cases}. \qquad \text{Eq.(8)}
$$

For example, for f_A we have

$$\frac{\partial f_A}{\partial \bar{\delta}_x} = \frac{1}{Den^2}\left[\left[\frac{\partial \Delta_M}{\partial \bar{\delta}_x}(\Phi_C - \Lambda_C) - \frac{\partial \Delta_C}{\partial \bar{\delta}_x}(\Phi_M - \Lambda_M)\right] Den - \left[\frac{\partial \Delta_A}{\partial \bar{\delta}_x}(\Phi_C - \Lambda_C) - \frac{\partial \Delta_C}{\partial \bar{\delta}_x}(\Phi_A - \Lambda_A)\right] Num\right].$$
$$\frac{\partial f_A}{\partial \bar{\lambda}_x} = \frac{1}{Den^2}\left[\frac{\partial \Lambda_C}{\partial \bar{\lambda}_x}(\Phi_M - \Delta_M) - \frac{\partial \Lambda_M}{\partial \bar{\lambda}_x}(\Phi_C - \Delta_C)\right] Den - \left[\frac{\partial \Lambda_C}{\partial \bar{\lambda}_x}(\Phi_A - \Delta_A) - \frac{\partial \Lambda_A}{\partial \bar{\lambda}_x}(\Phi_C - \Delta_C)\right] Num\Bigg].$$
$$\frac{\partial f_A}{\partial \bar{\phi}_x} = \frac{1}{Den^2}\left[\left[\frac{\partial \Phi_M}{\partial \bar{\phi}_x}(\Lambda_C - \Delta_C) - \frac{\partial \Phi_C}{\partial \bar{\phi}_x}(\Lambda_M - \Delta_M)\right] Den - \left[\frac{\partial \Phi_A}{\partial \bar{\phi}_x}(\Lambda_C - \Delta_C) - \frac{\partial \Phi_C}{\partial \bar{\phi}_x}(\Lambda_A - \Delta_A)\right] Num\right].$$
Eq.(9)

Using Eq. (9), the first-order Taylor series approximation (Taylor, 1982) for the variance of f_A evaluated at the mean can be calculated ~~(Taylor, 1982)~~ by:

$$\sigma_{f_A}^2 = \sum_x \left(\frac{\partial f_A}{\partial \bar{\delta}_x}\right)^2 \sigma_{\bar{\delta}_x}^2 + \sum_x \left(\frac{\partial f_A}{\partial \bar{\lambda}_x}\right)^2 \sigma_{\bar{\lambda}_x}^2 + \sum_x \left(\frac{\partial f_A}{\partial \bar{\phi}_x}\right)^2 \sigma_{\bar{\phi}_x}^2 = \sum_y \sum_x \left(\frac{\partial f_A}{\partial y_x}\right)^2 \sigma_{y_x}^2.$$
Eq.(10)

To calculate γ_A (the Satterthwaite (1946) approximation for the degrees of freedom), we define $f_{Ay_x} = c_A \left(\frac{\partial f_A}{\partial y_x}\right)^2$. In this case, we get:

$$\gamma_A = \frac{\left(\sum_y \sum_x f_{Ay_x} \sigma_{y_x}^2\right)^2}{\sum_y \sum_x \frac{\left(f_{Ay_x} \sigma_{y_x}^2\right)^2}{n_{y_x} - 1}}.$$
Eq.(11)

Note that whatever the value of c_A is, Eq. (11) leads to:

$$\gamma_A = \frac{\left(\sum_y \sum_x \left(\frac{\partial f_A}{\partial y_x}\right)^2 \sigma_{y_x}^2\right)^2}{\sum_y \sum_x \frac{\left(\left(\frac{\partial f_A}{\partial y_x}\right)^2 \sigma_{y_x}^2\right)^2}{n_{y_x} - 1}}$$

and if we set $f_{Ay_x}^* = \left(\frac{\partial f_A}{\partial y_x}\right)^2$ then the numerator of the last equation can be replaced by $\left(\sigma_{f_A}^2\right)^2$. In other words, we can use Eq. (10) and the derivatives (9) to estimate the value of γ_A resulting in $f_{Ay_x} = c_A f_{Ay_x}^*$. Of course, it is required that c_A is constant w.r.t. y_x. Then,

$$\gamma_A = \frac{\left(\sigma_{f_A}^2\right)^2}{\sum_y \sum_x \frac{\left(\left(\frac{\partial f_A}{\partial y_x}\right)^2 \sigma_{y_x}^2\right)^2}{n_{y_x} - 1}} \qquad \text{Eq.(12)}$$

Let $w \in \mathcal{C} \setminus \{A\}$. The first-order Taylor series approximation for the variance of f_w, can be calculated by (as above):

$$\sigma_{f_w}^2 = \sum_x \left(\frac{\partial f_w}{\partial \bar{\delta}_x}\right)^2 \sigma_{\bar{\delta}_x}^2 + \sum_x \left(\frac{\partial f_w}{\partial \bar{\lambda}_x}\right)^2 \sigma_{\bar{\lambda}_x}^2 + \sum_x \left(\frac{\partial f_w}{\partial \bar{\phi}_x}\right)^2 \sigma_{\bar{\phi}_x}^2 = \sum_y \sum_x \left(\frac{\partial f_w}{\partial y_x}\right)^2 \sigma_{y_x}^2. \qquad \text{Eq.(13)}$$

If we construct γ_w as γ_A, we get:

$$\gamma_w = \frac{\left(\sum_y \sum_x f_{w y_x}^* \sigma_{y_x}^2\right)^2}{\sum_y \sum_x \frac{\left(f_{w y_x}^* \sigma_{y_x}^2\right)^2}{n_{y_x} - 1}}$$

where $f_{w_{y_x}} = c_w f_{w_{y_x}}^*$ and $f_{w_{y_x}}^* = \left(\frac{\partial f_w}{\partial y_x}\right)^2$ with c_w constant w.r.t. y_x, then we finally get:

$$\gamma_w = \frac{\left(\sigma_{f_w}^2\right)^2}{\sum_y \sum_x \frac{\left(\left(\frac{\partial f_w}{\partial y_x}\right)^2 \sigma_{y_x}^2\right)^2}{n_{y_x} - 1}} \qquad \text{Eq.(14)}$$

The upper and lower confidence interval limits for each end-member fraction can be calculated using partial derivatives and the 95% confidence intervals constructed as:

$$f_w \pm t_{0.05\gamma_w} \sigma_{fw} \qquad \text{Eq.(15)}$$

Where $t_{0.05,\gamma}$ is the Student's t for α=0.05 (two-tailed) (Walpole et al., 2017) and γ degrees of freedom related with σ_{fw}.

3. Application

3.1. Study site and data

This methodology was tested using data from a high elevation (3,500 - 3,900 m a.s.l.) tropical catchment (7.53 km²) located in southern Ecuador (3°4′38″S, 79°15′30″O). The mean annual precipitation for this study site is 1,300 mm (Padrón et al., 2015), the mean annual discharge is 860 mm yr^{-1}. The catchment is of a volcanic origin and dominated by volcanic Histosol (24%) and Andosol (72%) soils (IUSS Working Group WRB, 2015), both with high percentage of organic matter content (450 and 310 g kg, respectively) (Quichimbo et al., 2012) and large water-holding capacities (Buytaert et al., 2006). Histosols are primarily located at the valleys and covered by cushion plants, while Andosol soils are predominated on the hillslopes under a cover of tussock grass. Nearlysaturated conditions of the riparian zone are observed year-round, and a spring is located in the north-western part of the catchment. Streamwater samples from 5 nested streams were collected weekly from March 2013 to April 2014 (n=~~270~~257) and at a higher frequency during experimental campaigns. ~~We also~~Additionally, ~~collected~~ bi-weekly water samples from 4 potential ~~water source~~ end-members: rainfall (RF), soil water from Andosols (AN) and Histosols (HS) and spring water (SW) (n ~ 30, for each end-member) were collected. The above-mentioned waters sources (RF, AN, HS and SW), were previously identified as end-members (Correa et al., 2017, 2019b)~~(Correa et al., 2017, 2018)~~ (Table 1). A multi-tracer (14 tracers) data set of conservative tracers was obtained from each water sample (Na, Mg, Al, Si, K, Ca, Rb, Sr, Ba, Ce, V, Y, Nd) at the Institute for Landscape Ecology and Resource Management of the Justus Liebig University using an ICP-MS (Agilent 7500ce, Agilent Technologies) and the electrical conductivity (EC) was measured in situ. More detailed information on the study site and data set can be found in Correa et al., (2017, 2019b)~~Correa et al. (2017, 2018)~~.

~~3.2.~~ Uncertainty estimation of water source contributions~~Source water uncertainty estimation~~

Using the classic EMMA approach (Christophersen and Hooper, 1992), end-members (source) and stream (mixture) data were projected into a three-dimensional space (Correa et al., 2019b)~~(Correa et al., 2018)~~ visualized in Figure 1. The resulting median and standard deviation of end-members and stream coordinates are shown in Table 1. Furthermore, Figure 2 shows the distribution of projected samples from individual end-members in the PCA coordinates.

The uncertainty of each of the four end-member contributions to the stream was determined using the above developed first-order Taylor series approximation from Eq. 14 (MatLab code in (Correa et al., 2019a)~~(Correa et al., 2019)~~. The variance for each end-member fraction ~~can~~ was ~~be~~ calculated ~~using partial derivatives and the 95% confidence intervals~~ (as recommended by Phillips and Gregg (2001)) ~~constructed~~ as $f_{EM1} \pm t_{0.05,\gamma}\, \sigma_{fEM1}$. The $t_{0.05,\gamma}$ depicts the Student's t for α=0.05 (two-tailed) and γ degrees of freedom. The γ degrees of freedom represents the Satterthwaite (1946) approximation for the related degrees of freedom with σ_{fEM1} and can be calculated as follows:

$$\gamma_{EM1} = \frac{\left(\sigma^2_{fEM1}\right)^2}{\sum_y \sum_x \frac{\left(\left(\frac{\partial f_{EM1}}{\partial y_x}\right)^2 \sigma^2_{y_x}\right)^2}{n_{y_x} - 1}} \qquad \text{Eq.(16)}$$

Note that Eq. (16) is an adaptation of Eq. (14) for this particular end-member configuration with x = EM1, EM2, EM3, EM4 and ~~SW~~M, y = $\bar{\delta}, \bar{\lambda}$ and $\bar{\phi}$, n= number of samples. The $\bar{\delta}, \bar{\lambda}$ and $\bar{\phi}$, ~~δ, λ and ϕ~~ represent the median of the projected water samples from end-members and stream in the principal components~~–~~U1, U2 and U3, respectively (U1 represents the principal components PC1, U2 PC2 and U3 and PC3). The f_{EM1} gives w the proportion of EM1 in ~~SW~~ M and σ^2_{fEM1}, the variances of the EM1. A similar procedure should be used for all end-members. The resulting uncertainty estimates for each source end-member are shown in Table ~~2~~5.

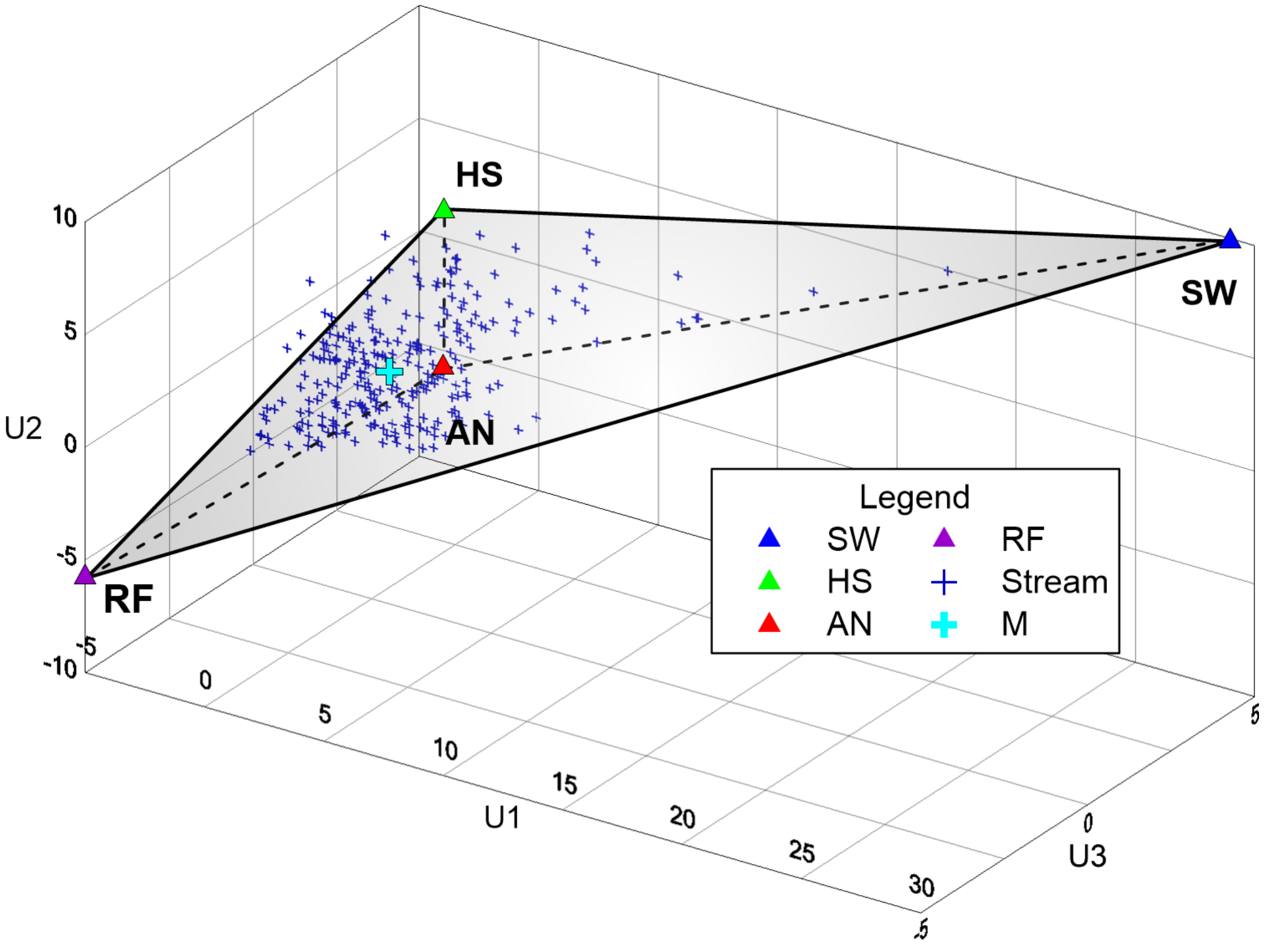

Figure 1. Three-dimensional mixing space generated using stream data, where the median of end-members are projected. U1 represents 59.6% of the variance, U2 19.7%, and U3 7.4% (From PCA); RF, rainfall; AN, Andosols; HS, Histosols; SW, spring water; M, median of stream data (mixture)

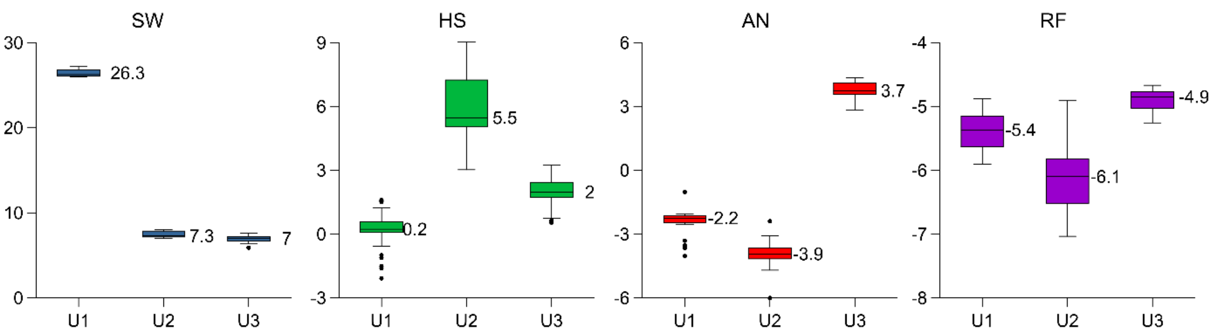

Figure 2. Boxplots of end-members projected in the three-dimensional mixing space for the study period 2013–2014, the Y-axis represents the coordinates of the mixing space and the X-axis the principal components U1, U2 and U3 (the central bar in the box represents the median; notches represent the 95% confidence intervals; whiskers 1.5 times the interquartile range and circles represent outliers). SW, spring water; HS, Histosol; AN, Andosol; RF, rainfall.

From the above-mentioned data set, we have generated 6 examples to assess~~nalyze~~ the sensitivity of the uncertainty calculation to the source sample size, the artificial inclusion of outliers (upper and lower extremes) and the increased standard deviations of the source datasets.

- The first example considers 50% of the samples from each source. The median, standard deviation and sample size are input data (Table 2) to calculate the uncertainty ranges (Table 6).

- The second considers the remaining 50% of samples and was similarly executed (Table 2).

- In the third example, outliers were artificially included at the upper positive end of data sets for each source at each coordinate, respectively. The outliers consisted of twice the maximum positive value of the observed data (Table 3).

- Using the same criteria, the negative extremes were included in the fourth example (Table 3).

- Sources affected by dispersed data clouds were taken into account by an increase in the standard deviation. We considered two cases, the first, in the example five, increasing three times the value of the standard deviation of the initial data set (Table 4) and finally, increasing the standard deviation five times for the sixth example (Table 4).

The results of this analysis are presented in Tables 6-8. In examples 1 and 2 the sample size reduction from 24 to 12 and 13 samples respectively (Table 6), had a minimal effect (less than 3%) on the calculation of the uncertainty ranges compared to the original complete set (Table 1). The fractions of source contributions did not experience changes. The inclusion of outliers affected the values of the medians at levels of the second decimal (Table 3) in relation to the median of the initial data (Table 2). However, the standard deviations increased in a range of 1.2 to 2.5 times the original value for AN and HS, and more for RF (2.5 to 10.5) and drastically for SW (4 to 20 times wider). These variations were reflected in the results of the calculation of uncertainties where the limits were extended for all existing cases from 1% to 12% (Table 6) in relation to Table 5. Furthermore, the widening of the standard deviations to three and five times their initial values resulted in an increase in the range of uncertainty between 2% and 22% for the first case and between 5% and 37% for the second case. For the latter, the minimum limit of the uncertainty range was reached in all the reported cases. The large number of samples used in these exercises were reflected in high degrees of freedom.

4. Summary and remarks

Our methodology developed to calculate the contribution of sources to the mixture and its associated uncertainty (based on multiple tracer sets) has been shown to be effective in real application cases. The robustness of the method is reflected in the fact that the calculations of the uncertainty ranges of multiple source contributions to a mixture do not experience significant changes with sample size reduction or inclusion of outliers. Rather, it shows marginally different results by incorporating standard deviations from widely dispersed data.

The simplicity of the methodology, based on Phillips and Gregg, (2001) combined with EMMA applications (Hooper, 2003) presents high potential for use as an alternative method to the simple sum of analytical errors (Uhlenbrook and Hoeg, 2003) or the Bayesian approach (Parnell et al., 2010; Stock et al., 2018). We provide a tool to help the community that has reported that a greater number of sources contribution and (common 2 or 3) the related uncertainty.is needed for a more complete conceptualization of the mixing processes (Iwasaki et al., 2015).

~~t~~The MatLab code provided and the illustrative examples facilitate~~s~~ ~~its~~ the understanding of the methodology and promote future scientific applications. We are confident that the use of this methodology will help the scientific community that is increasingly using large tracer sets in its research to obtain robust results.

5. Code and data availability

A MatLab code to calculate the fractions of end-members contribution to the mixture and their associated uncertainties is freely available in *https://zenodo.org/record/2649201*. As well as input data (used in this study) as an example for the code run and an instruction note.

6. Author contribution

AC and CB conceptualized the methodology. AC was responsible for the data collection and analysis. DO AC programmed and evaluated the MatLab code with collected data. AC wrote the manuscript with contributions from all co-authors.

7. Competing interests

The authors declare that they have no conflict of interest

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Table 1. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional space for the study period 2013–2014.

End-member		Coordinates* U1	U2	U3	Naming in equations
SW (n = 25)	median	26,25	7,29	7,00	A
	std.dev.	0,46	0,36	0,39	
HS (n = 33)	median	0,23	5,48	1,97	B
	std.dev.	0,85	1,29	0,69	
AN (n = 37)	median	-2,24	-3,93	3,71	C
	std.dev.	0,55	0,58	0,45	
RF (n = 36)	median	-5,38	-6,10	-4,84	D
	std.dev.	0,27	0,56	0,15	
Stream (n = 257)	median	-0,61	-1,04	0,94	M
	std.dev.	2,06	1,10	0,66	

* Coordinates of end-members and stream (mixture) medians in three-dimensional space (U1, U2 and U3). n represents the sample size.

Table 2. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional considering 50% of the data sets

Naming in equations		1) End member	Coordinates* U1	U2	U3	2) End member	Coordinates* U1	U2	U3
A	median	SW	26.18	7.29	6.66	SW	26.28	7.29	7.1
	std.dev.	(n = 12)	0.34	0.39	0.48	(n = 13)	0.51	0.36	0.21
B	median	HS	0.23	5.41	1.87	HS	0.28	5.9	2.26
	std.dev.	(n = 17)	0.74	1.19	0.52	(n = 17)	0.96	1.33	0.74
C	median	AN	-2.37	-3.93	3.69	AN	-2.2	-3.94	3.89
	std.dev.	(n = 19)	0.59	0.4	0.49	(n = 19)	0.46	0.73	0.41
D	median	RF	-5.37	-6.26	-4.78	RF	-5.35	-5.99	-5.01
	std.dev.	(n = 18)	0.26	0.58	0.07	(n = 18)	0.28	0.53	0.15
M	median	Stream	-0,61	-1,04	0,94	Stream	-0,61	-1,04	0,94
	std.dev.	(n = 257)	2,06	1,10	0,66	(n = 257)	2,06	1,10	0,66

The example 1) considers the initial 50% and 2) the remaining 50% of the sample sets.* Coordinates of end-members and stream (mixture) medians in three-dimensional space (U1, U2 and U3). n represents the sample size.

Table 3. Median and standard deviation (std.dev.) of end-members and stream projected in three-dimensional including artificial outliers

Naming in equations		3) End member	Coordinates* U1	U2	U3	4) End member	Coordinates* U1	U2	U3
A	median	SW	26.25	7.3	7.02	SW	26.21	7.29	6.95
	std.dev.	(n = 26)	5.51	1.73	1.68	(n = 26)	10.28	2.87	2.54
B	median	HS	0.27	5.47	1.98	HS	0.23	5.45	1.97
	std.dev.	(n = 34)	0.99	2.45	1.03	(n = 34)	1.12	1.99	0.8
C	median	AN	-2.24	-3.92	3.79	AN	-2.26	-3.95	3.74
	std.dev.	(n = 38)	0.78	1.17	0.92	(n = 38)	1.07	1.43	1.15

D	median	RF	-5.36	-6.08	-4.84		RF	-5.37	-6.11	-4.86
	std.dev.	(n = 37)	1.7	1.89	1.58		(n = 37)	1.09	1.42	0.94
M	median	Stream	-0,61	-1,04	0,94		Stream	-0,61	-1,04	0,94
	std.dev.	(n = 257)	2,06	1,10	0,66		(n = 257)	2,06	1,10	0,66

The example 3) considers outliers included at the positive extreme of the dataset of each source and 4) outliers included at the negative extreme.* Coordinates of end-members and stream (mixture) medians in three-dimensional space (U1, U2 and U3). n represents the sample size.

Table 4. Median and enlarged standard deviation (std.dev.) of end-members and stream projected in three-dimensional

Naming in equations		5)	End member	Coordinates* U1	U2	U3	6)	End member	Coordinates* U1	U2	U3
A	median		SW	26,25	7,29	7,00		SW	26,25	7,29	7,00
	std.dev.		(n = 25)	1.39	1.07	1.19		(n = 25)	2.32	1.78	1.99
B	median		HS	0,23	5,48	1,97		HS	0,23	5,48	1,97
	std.dev.		(n = 33)	2.56	3.87	2.06		(n = 33)	4.27	6.45	3.43
C	median		AN	-2,24	-3,93	3,71		AN	-2,24	-3,93	3,71
	std.dev.		(n = 37)	1.65	1.73	1.34		(n = 37)	2.75	2.88	2.24
D	median		RF	-5,38	-6,10	-4,84		RF	-5,38	-6,10	-4,84
	std.dev.		(n = 36)	0.8	1.69	0.46		(n = 36)	1.34	2.81	0.77
M	median		Stream	-0,61	-1,04	0,94		Stream	-0,61	-1,04	0,94
	std.dev.		(n = 257)	2,06	1,10	0,66		(n = 257)	2,06	1,10	0,66

The example 5) considers 3-times the standard deviation of the original data set and 6) 5-times the standard deviation of the original data set.* Coordinates of end-members and stream (mixture) medians in three-dimensional space (U1, U2 and U3). n represents the sample size.

Table ~~2~~5. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom calculated for the study period 2013–2014~~.~~

	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution	0.06	0.3	0.35	0.29
Upper 95% confidence limit	0.21	0.57	0.58	0.46
Lower 95% confidence limit	0.00	0.03	0.12	0.12
Degrees of freedom	291	536	749	628

Table 6. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed considering 50% of the data sets

	1)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	2)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.28		0.06	0.28	0.35	0.3
Upper 95% confidence limit		0.21	0.57	0.58	0.45		0.21	0.55	0.58	0.46
Lower 95% confidence limit		0.00	0.03	0.12	0.11		0.00	0.02	0.12	0.14
Degrees of freedom		289	493	676	589		288	491	679	537

The example 1) was computed considering the initial 50% and 2) the remaining 50% of the sample sets.

Table 7. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed after including artificial outliers

	3)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	4)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.29		0.06	0.3	0.35	0.29
Upper 95% confidence limit		0.22	0.62	0.64	0.5		0.22	0.61	0.63	0.49
Lower 95% confidence limit		0.00	0.00	0.06	0.08		0.00	0.00	0.07	0.08
Degrees of freedom		350	448	640	529		353	554	757	621

The example 3) was computed after including outliers at the positive extreme of the dataset and 4) including outliers at the negative extreme.

Table 8. Uncertainty of individual end-member contributions to the stream and Satterthwaite (1946) approximation for the degrees of freedom computed with enlarged standard deviations

	5)	EM1 SW	EM2 HS	EM3 AN	EM4 RF	6)	EM1 SW	EM2 HS	EM3 AN	EM4 RF
Fraction of end-members contribution		0.06	0.3	0.35	0.29		0.06	0.3	0.35	0.29
Upper 95% confidence limit		0.23	0.68	0.69	0.52		0.26	0.83	0.83	0.61
Lower 95% confidence limit		0.00	0.00	0.01	0.05		0.00	0.00	0.00	0.00
Degrees of freedom		372	225	362	312		335	122	211	172

The example 5) was computed considering 3-times the standard deviation of the original data set and 6) 5-times the standard deviation of the original data set.