Toolbox for accurate estimation and validation of PMF solutions in PM source apportionment

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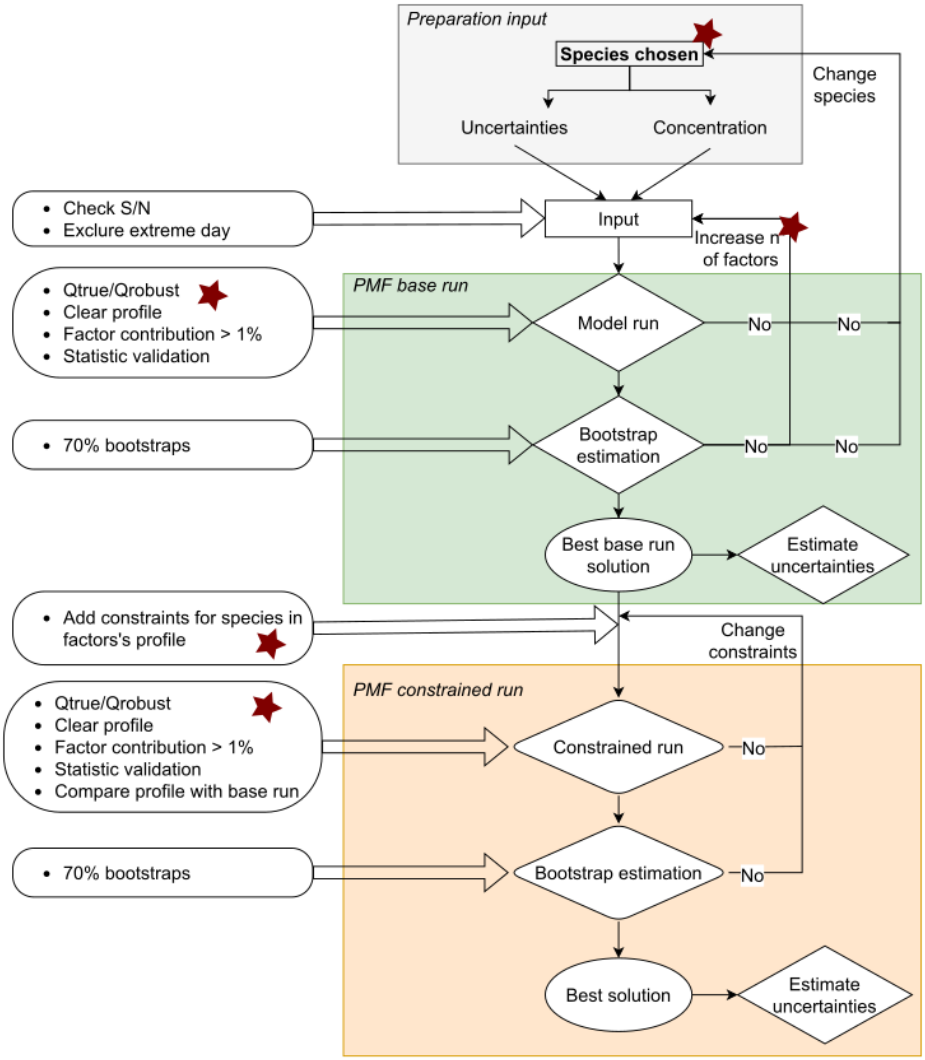

Figure S 1. Implementation for PMF. Red stars represent the steps including the subjectivity.

Table S 1. PMF input of classical solution and test 1, 2, 3, 4 (apply for database in Arrest and Ailly)

Test	Input variables
Basic solution	PM10 (total variable), OC*, EC Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, MSA Polyols (Arabitol + Mannitol) Levoglucosan, Mannosan 3-MBTCA Al, As, Ba, Cd, Co, Cu, Fe, Mn, Ni, Pb, Rb, Se, Sr, Ti, V, Zn
Test 1	Basic solution : Replace OC* by OC
Test 2	Basic solution : Remove Mannosan
Test 3	Basic solution : Repalce Polyols by Arabitol and Mannitol
Test 4	Basic solution + Oxalate + HULIS

Table S 2. PMF input of initial solution and test 5, 6, 7, 8, 9, 10, 11 (apply for the database in Grenoble)

Test	Input variables
Initial solution	PM10 (total variable), OC*, EC Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, MSA Polyols (Arabitol + Mannitol) Levoglucosan, Mannosan 3-MBTCA Al, As, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Rb, Sb, Se, Sn, Ti, V, Zn
Test 5	Initial solution + 2-MT1
Test 6	Initial solution + 2-MT2
Test 7	Initial solution + 2-MT3
Test 8	Initial solution + 2-MT4
Test 9	Initial solution + 2-MT5
Test 10	Initial solution + 2-MT3 + Fake 1
Test 11	Initial solution + 2-MT3 + Fake 1 + Fake 2

Table S 3. Ratio of chemicals in a source adopted form the references

Source	Ratio	Accepted range	Type	Reference
Biomass burning	Levoglucosan/K+	0.51 - 0.15	Crop residuals	(Li et al., 2021)
	Levoglucosan/Mannosan	>40	Crop residues	(Fu et al., 2012)
	Levoglucosan/Mannosan	15-25	Hardwood	(Fine et al., 2002)
	Levoglucosan/Mannosan	3-10	Softwood	(Fine et al., 2002)
	Levoglucosan/Mannosan	2.5	Charred pine wood	(Otto et al., 2006)
	Levoglucosan/Mannosan	0.3	Charred pine cone	(Otto et al., 2006)
	Levoglucosan/Mannosan	4.8 – 5.6	Forest fire smoke	(Ward et al., 2006)
	Levoglucosan/Mannosan	2-33.3	Grasses	(Oros et al., 2006)
	Levoglucosan/Mannosan	4	Green hardwood litter	(Medeiros and Simoneit, 2008)

	Levoglucosan/Mannosan	3.6	Green softwood litter	(Medeiros and Simoneit, 2008)
	Levoglucosan / OC	0.135	Residential wood burning	(Schauer et al., 2001)
	Levoglucosan / OC	0.010 - 0.334	Residential wood burning	(Fine et al., 2001)
	Levoglucosan / OC	0.043	Burn enclosure	(Hays et al., 2002)
	Levoglucosan / OC	0.045 ± 0.069	Sticks	(Mazzoleni et al., 2007)
	Levoglucosan / OC	0.043 ± 0.034	Needles	(Mazzoleni et al., 2007)
	Levoglucosan / OC	7 - 17	Wood burning	(Fine et al., 2002)
	OC/EC	3-10.6	BB burning	(Fine et al., 2001; Li et al., 2021)
Dust	OC/EC	2.3–17.7	Road dust	(Amato et al., 2009)
	Si/Al	1.4-3.19	Non-urban dust	(Rahn, 1976)
	Si/Al	2-4.63	Urban dust	(Rahn, 1976)
	La/Ce	0.4-0.6	Naturals dust	(Moreno et al., 2010)
	V/Ni	12	Soil dust	(Viana et al., 2009)
	V/Ni	32	African dust	(Viana et al., 2009)
	Fe/Al	0.51-0.73	Saharan dust	(Guieu et al., 2002)
	Ti/Ca	0.012-0.018	Normal day – Dust event	(Nicolás et al., 2008)
	Ti/Fe	0.085-0.105	Normal day – Dust event	(Nicolás et al., 2008)
	Al/Ca	0.251-0.253	Normal day – Dust event	(Nicolás et al., 2008)
	Fe/Ca	0.14-0.17	Normal day – Dust event	(Nicolás et al., 2008)
Traffic	Cu/Sb	7 - 9	Brake wear urban site	(Amato et al., 2009)
	OC/EC	0.28-0.52	Diesel	(Amato et al., 2011)
	OC/EC	2.2	Gasoline	(Na et al., 2004)
	OC/EC	1.34-3.7	Road traffic	(Giugliano et al., 2005)
	Sn/Sb	1.49-1.5	Road traffic	(Amato et al., 2011)
	Cu/Sn	4.7-5.4	Road traffic	(Amato et al., 2011)
	Cu/Sb	6.8-8.0	Road traffic	(Amato et al., 2011)
	Fe/Sn	93-109	Road traffic	(Amato et al., 2011)
	Fe/Sb	137-158	Road traffic	(Amato et al., 2011)
	Fe/Cu	17-29	Road traffic	(Amato et al., 2011)
	Sb/Cd	5.0	Brake wear urban site	(Thorpe and Harrison, 2008)
HFO	V/Ni	2.5-5	Shipping	(Viana et al., 2009)
	La/Ce	>1	Petrolium coke	(Moreno et al., 2010)
	V/EC	<2	Shipping	(Viana et al., 2009)
Sea salt	Na^+ / Cl^-	1.8	Fresh sea salt	(Marenco et al., 2007)
	Ca/Na	0.04	Sea salt	(Waked et al., 2014)
	Mg/Na	0.12	Sea salt	(Waked et al., 2014)

Primary biogenic	OC/Polyols	0.03 – 0.08	Plant debris/ Fungi	(Bauer et al., 2002; Yttri et al., 2011)
	OC/(Arabitol+Mannitol)	0.11 ± 0.12	Plant debris/ Fungi	(Weber et al., 2019)
MSA rich	MSA/OC	0.4 ± 0.3	MSA rich	(Weber et al., 2019)
Secondary oxidation	3-MBTCA/OC	0.012 ± 0.002	Alpha-pinene SOA	(Borlaza et al., 2021)
Nitrate rich	NO_3^-/NH_4^+	0.21-0.32	Secondary nitrate	(Kai et al., 2007)
	NO_3^-/NH_4^+	0.6	Molar ratio	(Heo et al., 2013)
Sulfate rich	SO_4^{2-}/NH_4^+	0.22-0.3	Secondary sulfate	(Kai et al., 2007)
	NO_3^-/NH_4^+	0.4	Molar ratio	(Heo et al., 2013)

Table S 4. SPECIEUROPE database description

	Name	Specie	Particle Size	Country
	2STROKE, AGET, AMNITR, AMSULF, APP_10, APP_2.5, ASPHALTS, ASVTVEL, AgedSea, Aluminium_10, Aluminium_2.5, Ash_10, Ash_2.5, Asphalt, Asphalt_10, Asphalt_2.5, BMTE, BRAKE, BRONZESM, BUSDIES, Beech, Bell_10, Bell_2.5, Biomass Burning, Biomass burning, BkgRawSoil, BkgSoil, Blast furnace, BrakeTyre, BrakesC_10, BrakesC_2.5, Briks_10, Briks_2.5, Briquettes, Burn1_2, CAO, CBURN-B, CBURN-P, CEGASOL, CEMENFF, CEMENRM, CEMENT, CEMENTP, CGASCAR, CaCl2, Cast iron, CementMill_10, CementMill_2.5, Cement_10, Cement_2.5, Ceramic_10, Ceramic_2.5, CoalMill_10, CoalMill_2.5, Coal_10, Coal_2.5, Contruction, Copper_10, Copper_2.5, Crustal, CrustalDust, Crustal_PP, Crustalresusp, DIESBUS1, DIESBUS3, DIESBUS4, EAF-Slag_10, EAF-Slag_2.5, EAF-Steel_10, EAF-Steel_2.5, EUSAAR2_10, EUSAAR2_2.5…etc	Al, Sb, As, Cd, Ca, Cr, Cu, Fe, Pb, Mg, Mn, Mo, Ni, NO3-, OC, K, Rb, Si, Na, Sr, SO4=, Sn, Ti, V, Zn, NH4+, Cl, EC, BAPY, BEPY, BGHI, CHRY, CORO, INCD, levg, BbF, BkF, DBAHA, Ba, Cl-,S , Na+, Br, K+, Ca2+, nan, BAAN, FLUORA, PYRE, Ga, Se, Zr, PERY, CPCDP, ACRI, Co, Ag, Tl, U, Li, Th, Bi, TC, PO4, CO3=, Nb, Al2O3, Y, Be, Sc, Ta, W, Cs, Ge, O2Si, REE, La, PAH, Te, galc, mann, P, Ce, Sm, Eu, BZAN, RETE, abac, deca, doca, ecos, hexa, laua, myra, ndec, pala, stea, syri, teco, trco, N_EICO, S192, Mg2+, HULIS, MA, azea, pdec, tdec, BjF, ACETO, BENZE, In, N_DODE, NAPHTH, Pd, TOLUE, ACNA, FLUORE, PHEN, N_HEXD, N_NOND, N_OCTD, N_TETD, Nd, Pr, Hf, Dy, B, Gd, Er, C29H50, C31H54, Hg, WSOC	PM10, TSP, PM2.5, PMraw, PM50, PM1, PM	Italy, Poland, Spain, Sweden, France, Denmark, Greece, Turkey, Austria, Germany, Vancouver, US, Portugal, Western Mediterranean sea
Total	291	134	7	15

Table S 5. IGE database (use for this study) description

	Name	Specie	Particle Size	Country/Site
	MSA rich, Dust, Biomass burning, Sea salt, Aged sea salt, Industrial, HFO, Nitrate rich, Traffic, Secondary oxidation, Primary biogenic, id, Seasalt, Aged seasalt, Road traffic, Secondary biogenic, Sulfate rich/HFO, Industries, Sulfate rich, Road salt, MSA-rich, Primary traffic, Sulfate-rich, Sea/road salt, Secondary biogenic oxidation, Nitrate-rich, Mineral dust, Fresh seasalt, Sea-road salt, Vanadium rich, Secondary biogenic/HFO, Cadmium rich, HFO (stainless)	OC*, EC, Cl-, NO3-, SO42-, Na+, NH4+, K+, Mg2+, Ca2+, MSA, Levoglucosan, Mannosan, Polyols, 3-MBTCA, Cellulose, Phthalic, Pinic Al, As, Ba, Cd, Co, Cu, Fe, Mn, Ni, Pb, Rb, Se, Sr, Ti, V, Zn, Cr, La, Mo, Sb, Sn, Cs, Ce, Li, Zr, Tl	PM10	French sites
Total	33	44	1	18

Table S 6. Formula to calculate uncertainties

Specie	Calculation by	Formula
OC*, EC, PM10	Fixed percentage	10%
Sample which has concentration < QL	Ratio of QL Polissar et al. (1998)	$\frac{5 \times QL}{6}$
The others	Gianini et al., 2012	$\sigma_{ij} = \sqrt{QL_j^2 + (CV_j \times x_{ij})^2 + \left(a \times x_{ij}\right)^2}$

With:

- QL : Quantification limit.
- CV : Coefficient of variation.
- a: Additional coefficient of variation
- x: Species concentration.

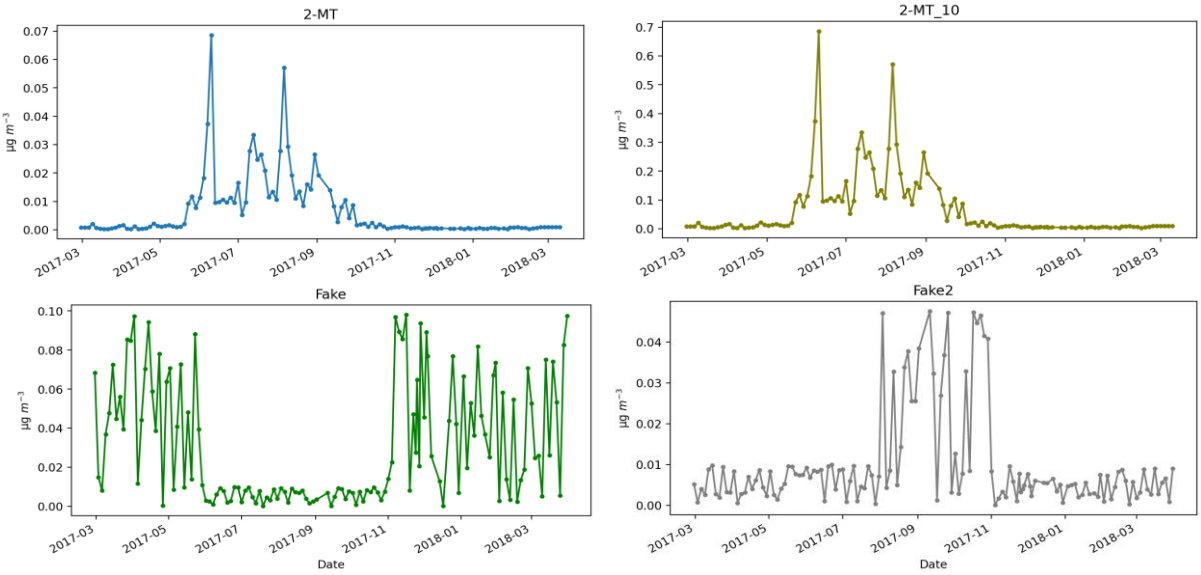

Figure S 2. Temporal evolution of 2-MT and Fake components

Table S 7. Bootstrap values of the base runs and constraint runs for Tests 1 to 4 at Ailly

	Basic		Test 1		Test 2		Test 3		Test 4	
	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint
Aged sea salt	100	100	99	99	100	100	99	98	99	100
Biomass burning	100	100	100	100	100	100	100	100	100	100
Dust	90	100	91	98	99	99	97	100	88	96
HFO	97	99	97	100	96	99	94	100	95	95
Industrial	87	100	85	100	70	95	76	100	70	100
MSA rich	100	100	99	100	100	100	99	100	100	100

Nitrate rich	100	100	100	100	100	100	100	100	100	100
Primary biogenic	79	99	77	100	88	99	95	100	88	100
Sea salt	100	100	100	100	100	100	100	100	100	100
Secondary oxidation	99	100	98	100	99	100	99	100	98	100
Traffic	88	100	86	100	95	100	84	100	86	100
R^2 PM_{obs} and PM_{pred}	0.99	0.99	0.99	0.99	0.99	0.99	0.99	0.99	0.99	0.99
Q_{true}/Q_{robust}	1.09	1.08	1.09	1.08	1.08	1.06	1.08	1.08	1.05	1.05
Q_{true}/Q_{expect}	2.47		2.45		2.50		2.30		2.71	

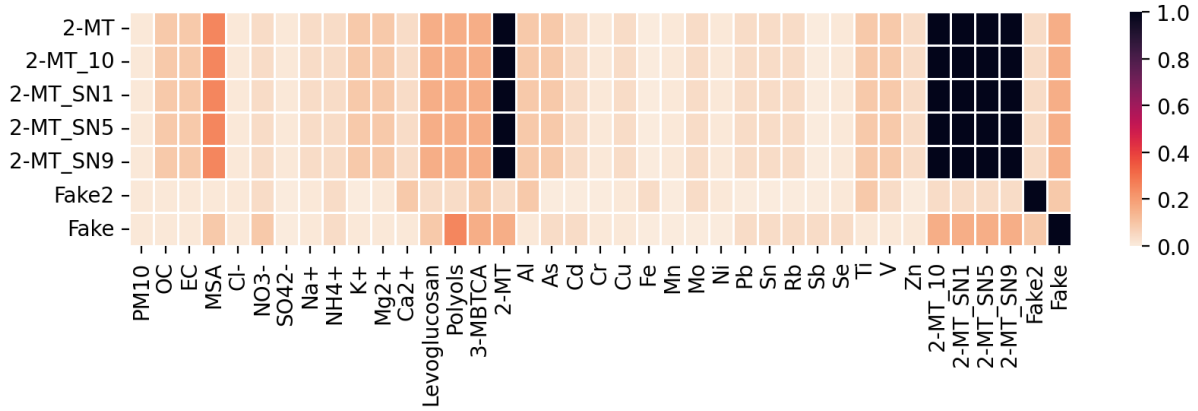

Figure S 3. Correlation determination of 2-MT and Fake components with the chemical included in the PMF (test 5 to test 11).

Table S 8. Constraint applied for Tests 5 to 11.

Factor	Element	Type	Applied for
Fake 2	Fake2	Pull Up Maximally	Tests 11
Fake 1	Fake	Pull Up Maximally	Test 10, 11
Primary biogenic	Polyols	Pull Up Maximally	Tests 5 to 11
Biomass burning	Levoglucosan	Pull Up Maximally	Tests 5 to 11
MSA rich	MSA	Pull Up Maximally	Tests 5 to 11
2-MT	2-MT	Pull Up Maximally	Test 6, 7, 8, 10, 11
Secondary oxidation	3-MBTCA	Pull Up Maximally	Tests 5 to 11
Traffic	Sn	Pull Up Maximally	Tests 5 to 11
Traffic	Sb	Pull Up Maximally	Tests 5 to 11

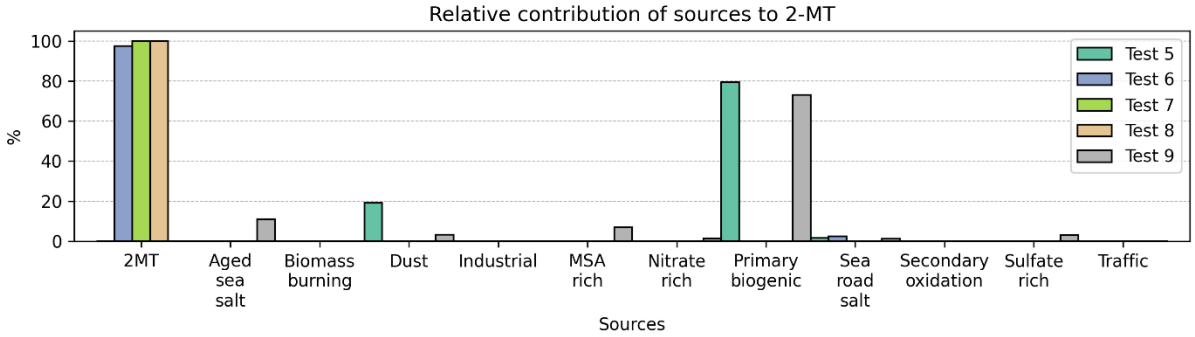

Figure S 4. Contribution of sources to 2-MT in test 5,6,7,8,9

Table S 9. R^2 and bootstrap values of the constraint run

	Test 5		Test 6		Test 7		Test 8		Test 9		Test 10		Test 11	
	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint	Base	Constraint
Aged sea salt	100	100	100	100	100	100	100	100	100	100		100	100	100
Biomass burning	100	100	99	100	99	100	100	100	100	100	100	100	100	100
Mineral dust	75	90	95	99	94	98	95	98	94	99	98	100	98	100
Industrial	100	100	100	97	100	100	100	100	100	100	98	100	100	100
MSA rich	95	100	95	100	96	100	98	100	98	100	100	100	100	100
Nitrate rich	100	100	100	100	100	100	100	100	100	100	100	100	100	100
Primary biogenic	95	95	96	100	84	100	72	100	91	100	86	100	86	100
Sea/road salt	100	100	100	100	100	100	100	100	100	100	100	100	100	100
Secondary oxidation	100	100		100	100	100	98	100	96	100	100	100	100	100
Sulfate rich	100	100	96	100	99	99	99	100	96	100	99	100	96	100
Traffic	95	100	98	100	99	100	98	100	100	100	99	100	99	100
2-MT	X	X	60	99	60	99	100	100	X	X	84	100	65	100
Fake 1	X	X	X	X	X	X	X	X	X	X	100	100	80	100
Fake 2	X	X	X	X	X	X	X	X	X	X	X	X	79	100
R^2 PM_{obs} and PM_{pred}	0.91	0.91	0.90	0.90	0.90	0.90	0.90	0.90	0.90	0.90	0.90	0.90	0 .90	0.90

Table S 10. DISP values ([5th, 95th]) of 2-MT, Fake 1, Fake 2 in the chemical profile that they contribute most.

	Test 5	Test 6	Test 7	Test 8	Test 9	Test 10	Test 11
2-MT	[0.027, 0.031]	[0.065, 0.073]	[0.063, 0.068]	[0.061, 0.062]	[0.001, 0.001]	[0.0063, 0.0068]	[0.0063, 0.0068]
Fake 1	X	X	X	X	X	[0.031,0 .034]	[0.030,0 .033]
Fake 2	X	X	X	X	X	X	[0.010, 0.013]

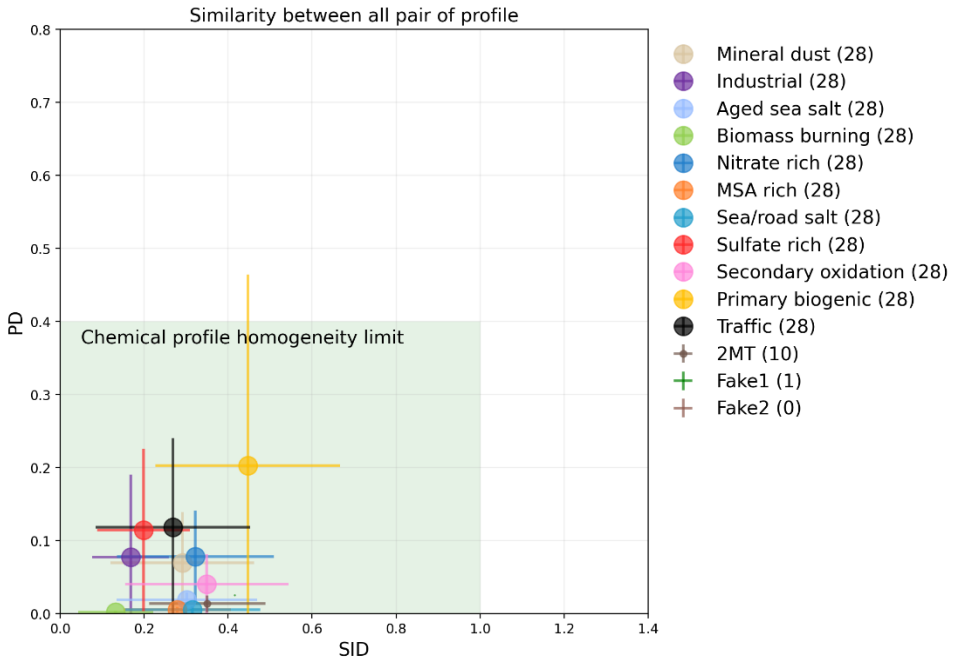

Figure S 5. Similarity the sources of test 5 to test 11. The shaded area denotes the limit of homogeneity. The error bar of each point represents the standard deviation of PD and SID of the comparison. The number between parentheses in the legend represents the number of all pairs used for the comparison.

S1. Tools development to evaluate the accuracy of PMF solution

Python package has been developed with many tools needed when performing a PMF study with series of off-line samples, in order to improve the efficiency of performing it (Figure S6). It includes some aspects described in this study (red dot frame) and some additional tools that can be implemented to reduce the subjectivity of the choices to be made. This package is composed of several sections, that extend the features included in EPA PMF5.0. These sections are: (1) input preparation and visualisation (2) validation and tests on PMF results and (3) visualisation of PMF final result.

Tools from section (1) support steps before running a PMF, including uncertainty calculation, data description, data curation (observation of temporal evolution, correlation, clustering between chemical species). It also allows an evaluation if a chemical can be a proper tracer, according to the conditions

presented in section 3.2. Tool (2) validates the PMF solution, using the statistical parameters such as: the R^2 between observed and predicted concentration, bootstrapping and displacement, Q_{true}/Q_{robust} and Q_{true} / Q_{expect} values. It further includes geochemical validation with tests on diagnostic ratios and chemical profiles, as **developed in sections 3.3**. Tool (3) is used to visualize the chemical profile and contribution of source exported from PMF result (for single site and multiple sites). Especially, the visualisation is also take into account the BS values and DIPS values for the chemical profile and contribution, which presented by Weber et al. (2019).

The package and its documentation are available on GitHub, which are free accessed and can be download at: https://github.com/DinhNgocThuyVy/PMF_tools.

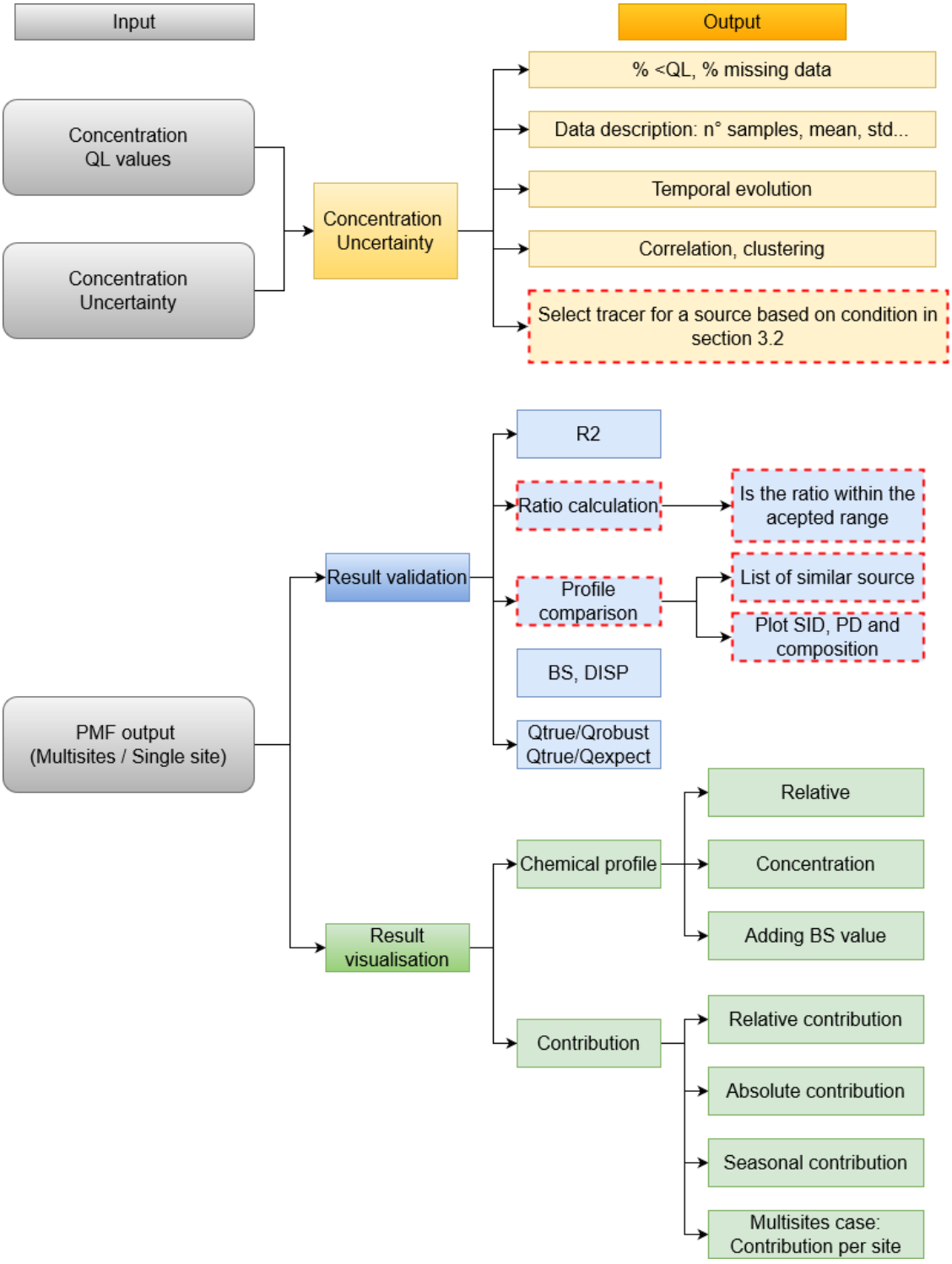

Figure S 6. Workflow of the tools. The dot red frame denotes the tools developed in this study.

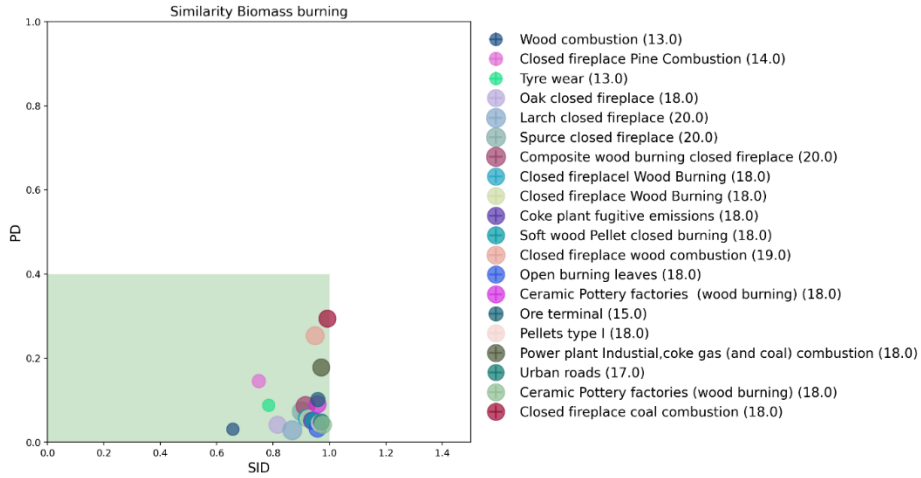

Figure S 7. Similarity of soucres SPECIEUROPE to BB in GRE-fr. The shaded area denotes the limit of homogeneity. The error bar of each point represents the standard deviation of PD and SID of the comparison. The number between parentheses in the legend represents the number of the chemical species used for the comparison.

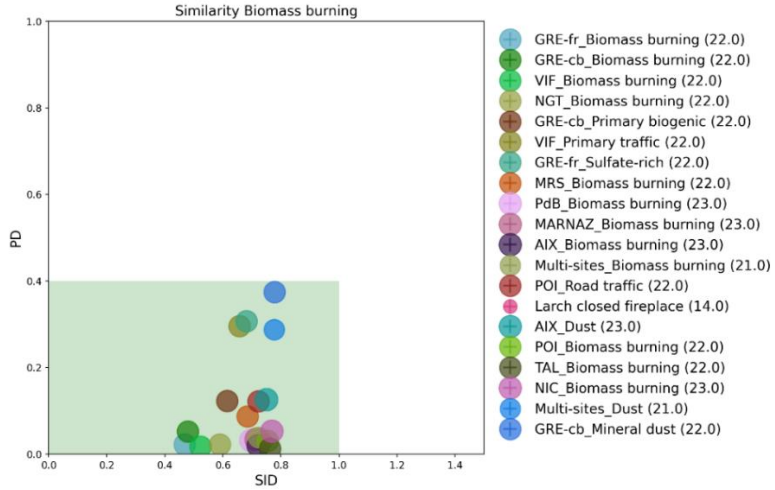

Figure S 8. Similarity between sources of SPECIEUROPE and IGE compare to GRE-fr. The shaded area denotes the limit of homogeneity. The error bar of each point represents the standard deviation of PD and SID of the comparison. The number between parentheses in the legend represents the number of the chemical species used for the comparison.

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