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Original article

## SPECIEUROPE: The European data base for PM source profiles



Denise Pernigotti\*, Claudio A. Belis\*, Luca Spanò

European Commission, Joint Research Centre, Institute for Environment and Sustainability, Air and Climate Unit, Via E. Fermi 2749, 21027 Ispra, VA, Italy

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### ABSTRACT

A new database of atmospheric particulate matter emission source profiles in Europe (SPECIEUROPE) developed in the framework of the Forum for air quality modeling in Europe (FAIRMODE, Working Group 3) is accessible at the website <http://source-apportionment.jrc.ec.europa.eu/Specieurope/index.aspx>. It contains the chemical composition of particulate matter emission sources reported in the scientific literature and reports drafted by competent authorities. The first release of SPECIEUROPE consists of 151 measured (original), 13 composite (merging different subcategories of similar sources), 6 calculated (from stoichiometric composition) and 39 derived (results of source apportionment studies) profiles. Each profile is related to one or more source categories or subcategories. The sources with the highest PM relative mass toxic pollutants such as PAHs are fuel oil burning, ship emissions, coke burning and wood burning. Heavy metals are most abundant in metal processing activities while halogens are mostly present in fertilizer production, coal burning and metallurgical sector. Anhydrosugars are only measured in biomass and wood burning source categories, because are markers for these categories. The alkaline earth metals are mostly present in road dust, cement production, soil dust and sometimes coal burning. Source categories like traffic and industrial, which contain heterogeneous subcategories, show the greatest internal variability.

The relationships between sources profiles were also explored using a cluster analysis approach based upon the Standardized Identity Distance (SID) indicator. The majority of profiles are allocated in 8 major clusters. Some of the clusters include profiles mainly from one source category (e.g. wood burning) while others, such as industrial source profiles, are more heterogeneous and spread over three different clusters.

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## 1. Introduction

Particulate matter (PM) source profiles (fingerprints) are the average relative chemical composition of the PM deriving from a pollution source, commonly expressed as the mass ratio between each species to the total PM. Local or regional source profiles are essential for PM source apportionment studies carried out with receptor models (RMs). These tools treat the monitored air quality data at a receptor site to estimate the sources of air pollution (Viana et al., 2008; Hopke and Cohen, 2011; Belis et al., 2013). In the chemical balance model (CMB) approach, the fingerprints are

required as input data (Watson et al., 1997). On the contrary, no a priori knowledge about pollution sources is required in multivariate factor analysis (MFA). In this case the output consists of factors that have to be interpreted in terms of real-world emission sources. The availability of source profiles representative of the sources in the study area is, therefore, essential as input for models using the CMB approach and as references to validate the attribution of factors to source categories in the MFA approach.

The United States EPA SPECIATE database of source profiles has been available since 1988 (Simon et al., 2010) and currently contains over 3000 entries. In Europe, the scarcity of local source profiles often represented a challenge for RMs studies in terms of both the identification of sources and the comparability between studies. Source profiles from SPECIATE are used in Europe when the sources are comparable. Nevertheless, certain sources (e.g. mineral dust and traffic exhaust) depend much on local variables like the geological substrate or the circulating fleet. The objectives of SPECIEUROPE are to fill the gap in the availability of input data for

\* Corresponding authors.

E-mail addresses: [denise.pernigotti@jrc.ec.europa.eu](mailto:denise.pernigotti@jrc.ec.europa.eu) (D. Pernigotti), [claudio.belis@jrc.ec.europa.eu](mailto:claudio.belis@jrc.ec.europa.eu) (C.A. Belis).

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source apportionment and to provide reference source profiles for the whole Europe.

The paper is divided in two parts, the first (section 2) an overview of the source profiles present in SPECIEUROPE, allocated in source categories on the basis of their name and description, is given. In the second part (section 3), the comparability of the profiles within and among source categories is assessed using cluster analysis.

## 2. Database description

In SPECIEUROPE (2015) the relative species' concentrations and their uncertainties are reported as the ratio of the mass to the PM total mass. Since the minimum detection limit is in general not known, values below the detection limit (BDL) are flagged as 'not detected'. Even though the uncertainty is important for the proper source allocation with RMs, it is not always reported in the studies where the source profiles were originally published. In addition, when present it is expressed in various forms: standard deviation, half range, full range or 95% confidence interval. In some cases, the given analytical uncertainty may be underestimated because it does not duly account for the sampling uncertainty.

Fingerprints reported in SPECIEUROPE are either 'O' original, 'C' composite (merging different subcategories of similar sources), 'T' theoretical (e.g. stoichiometric ratios) or 'D' derived (as from MFA or CMB outputs) and refer, with few exceptions, to the total mass of PM<sub>2.5</sub> and PM<sub>10</sub>.

### 2.1. The database architecture

The database was created under the SQL server database management system. The information is arranged in tables as follows:

- 1) Profiles' species relative concentrations, their uncertainties and the analytical technique used.
- 2) Single profile name and description.
- 3) Publication information (publications often contains more than one profile).
- 4) Information on source categories related to a single profile.

Ancillary tables store the codification system used for uncertainty methods, chemical families, chemical species, chemical analytical methods and source categories' description.

Each species is identified with a unique internal identification number (IDN), when possible corresponding to the one assigned in SPECIATE (Simon et al., 2010). The CAS number and the AIRBASE code (European Commission, 2014) are reported when available. Likewise, the source profiles are identified by a unique IDN, which should be reported whenever the profiles are used to guarantee an efficient and univocal identification throughout the literature. Source profiles attributed to a given source category are also allocated to the higher level categories containing it; i.e. if a fingerprint is attributed to the source category gasoline, it is also attributed to the more comprehensive source categories exhaust and traffic.

### 2.2. Overview of the current release

The first SPECIEUROPE (2015) release consists of 209 profiles 151 of which are original 'O', 39 derived 'D', 13 composite 'C' and 6 theoretical 'T'. With the exception of 10 ship profiles (Lu et al., 2006; Agrawal et al., 2008), all profiles are derived from European scientific papers or reports (Alastuey et al., 2006; Alleman et al., 2010; Amato et al., 2011; Argyropoulos et al., 2013; Bernardoni et al., 2011; Cesari et al., 2012; Colombi et al., 2010; Diesch et al., 2013; El Haddad et al., 2009; Larsen et al., 2008,

2012; Moldanová et al., 2009; Pey et al., 2013a,b; Pietrodangelo et al., 2013; Samara et al., 2003; Sánchez de la Campa et al., 2010; Schmidl et al., 2008a,b; Sjödin et al., 2010; Wahlin et al., 2006; Yatkin and Bayram, 2008). The marine aerosol profile is taken from Anderson (2015) and two deicing salt profiles are derived from product technical specifications (ESCO, 2015; Houska and TMR Consulting, 2015).

In this section are only discussed the 163 profiles deriving from measures ('O' and 'C'). Source categories (with their IDN) with at least one original or composite profile are reported in Table 1. The number of profiles (#prof), the average number of common species among profiles (#spec) and the number of publications used (#pub) for every source category are also reported. The most populated source categories, namely industry, traffic, and biomass burning, include subcategories. Among the more specific categories, those with the highest number of profiles are soil dust, wood burning and road dust. The remaining categories include profiles from at most four publications. It has to be noted that the ship exhaust is mainly composed of numerous profiles from a single publication (Agrawal et al., 2008). Deicing salt and marine aerosol have been excluded from the analysis because are not comparable

**Table 1**

SPECIEUROPE source categories are reported by IDN and name. The number of O and C profiles (#prof), the average number of common species among profiles (#spec) and the number of publications (#pub) are reported.

IDN	Source category name	#prof	#spec	#pub
1	Traffic	28	14.3	9
5	Road dust	15	14.2	8
20	Industrial	77	17.0	7
40	Biomass burning	24	20.8	6
10	Soil dust	20	14.8	6
41	Wood burning	18	23.8	6
30	Fuel oil burning	11	28.2	5
47	Closed fireplace	16	25.7	4
37	Ship exhaust	14	21.7	4
2	Exhaust	12	17.6	4
25	Cement	11	15.1	4
28	Power plant	10	19.5	4
34	Boiler	8	18.0	4
66	Deicing salt	6	2.2	4
31	Coal burning	12	20.5	3
21	Iron & steel prod.	7	16.0	3
32	Coke burning	6	24.9	3
12	Marine aerosol	3	5.7	3
29	Fertilizer prod.	9	29.3	2
22	Foundries	6	14.2	2
27	Ceramic	6	27.1	2
3	Diesel exhaust	5	19.3	2
4	Gasoline exhaust	4	20.0	2
24	Metal smelting	4	18.5	2
54	Hard wood burning	4	34.0	2
33	Natural gas burning	3	15.3	2
43	Pellet burning	3	19.7	2
53	Soft wood burning	3	26.3	2
44	Beech burning	2	15	2
46	Leaves burning	2	13	2
55	Open burning	2	13	2
14	Volcanic dust	2	16	1
35	Petrochemical	2	38	1
49	Olive oil burning	2	16	1
60	Second. inorg. Aer.	2	1	1
6	Tyre wear	1	8	1
7	Brake dust	1	17	1
23	Refineries	1	22	1
26	Incinerator	1	23	1
42	Pine burning	1	23	1
50	Oak burning	1	41	1
51	Spruce burning	1	77	1
52	Larch burning	1	41	1
61	Ammonium nitrate	1	2	1
62	Ammonium sulfate	1	2	1

to the other source profiles due to the limited number of species they contain.

Figs. 1 and 2 summarize the fingerprints for the most common SPECIEUROPE source categories. On the abscissa only the 43 most abundant species (as reported in Table 2 with relative mass over 0.008) are displayed, according their decreasing maximum relative mass. In certain profiles, the concentrations of Cl, Na, K, Ca and Mg are reported both as total and as ion. To prevent double counting of the mass of these species, only the higher of the two has been considered (e.g. the Calcium in Table 2). The Polycyclic aromatic

hydrocarbons (PAHs) have been aggregated in one single species in order to have more comparable values.

The box plots in Figs. 1 and 2 represent the statistical distribution of the chemical species in the profiles attributed to the same category. For example, there is a considerable variability in the organic carbon and lead content among the sources in the coke burning source category. The same can be seen in the chlorine, organic and elemental carbon relative concentrations of the coal burning source category, and the iron, nitrate and sulfate relative abundances of the profiles in the fertilizer production source

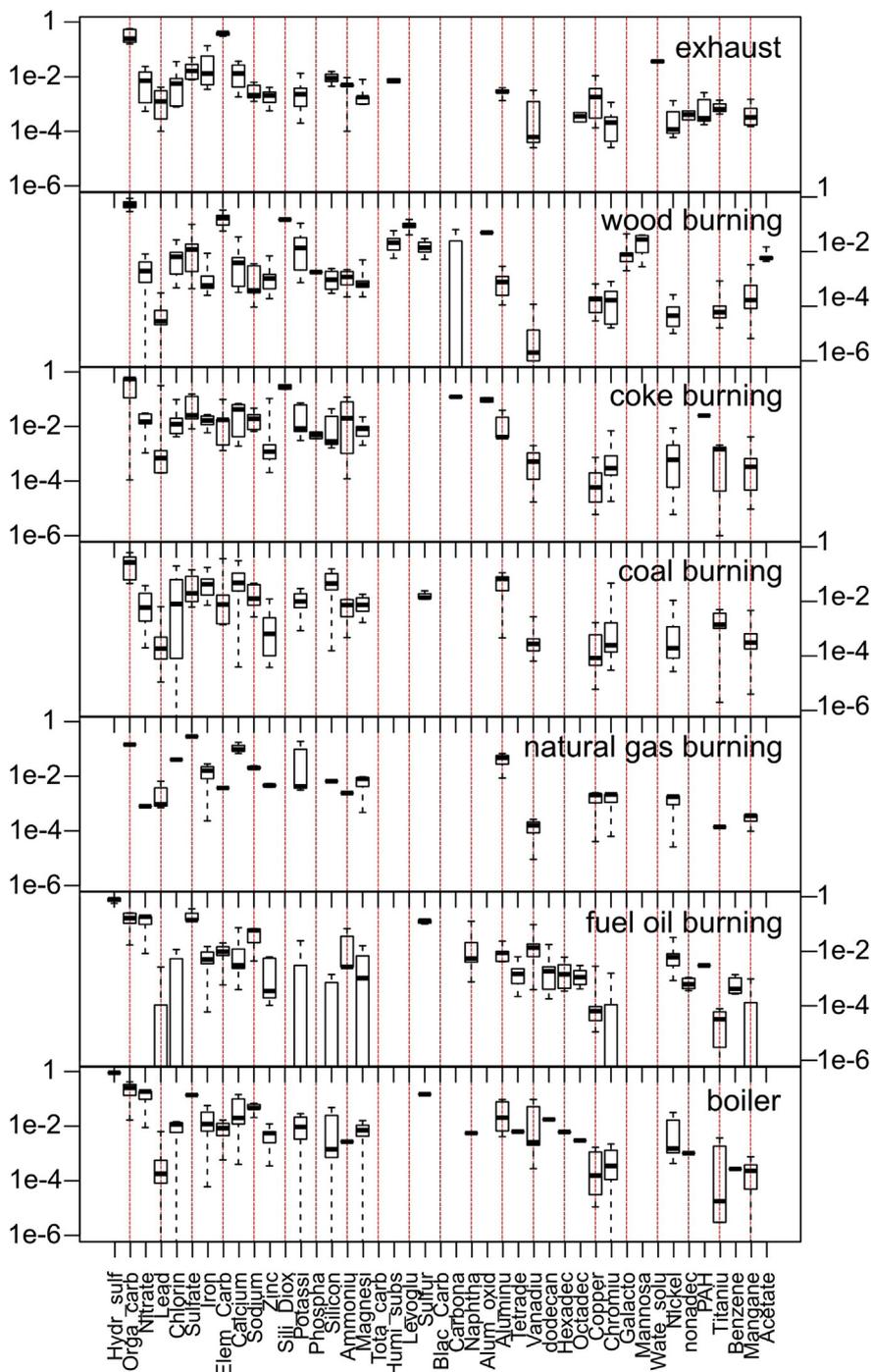
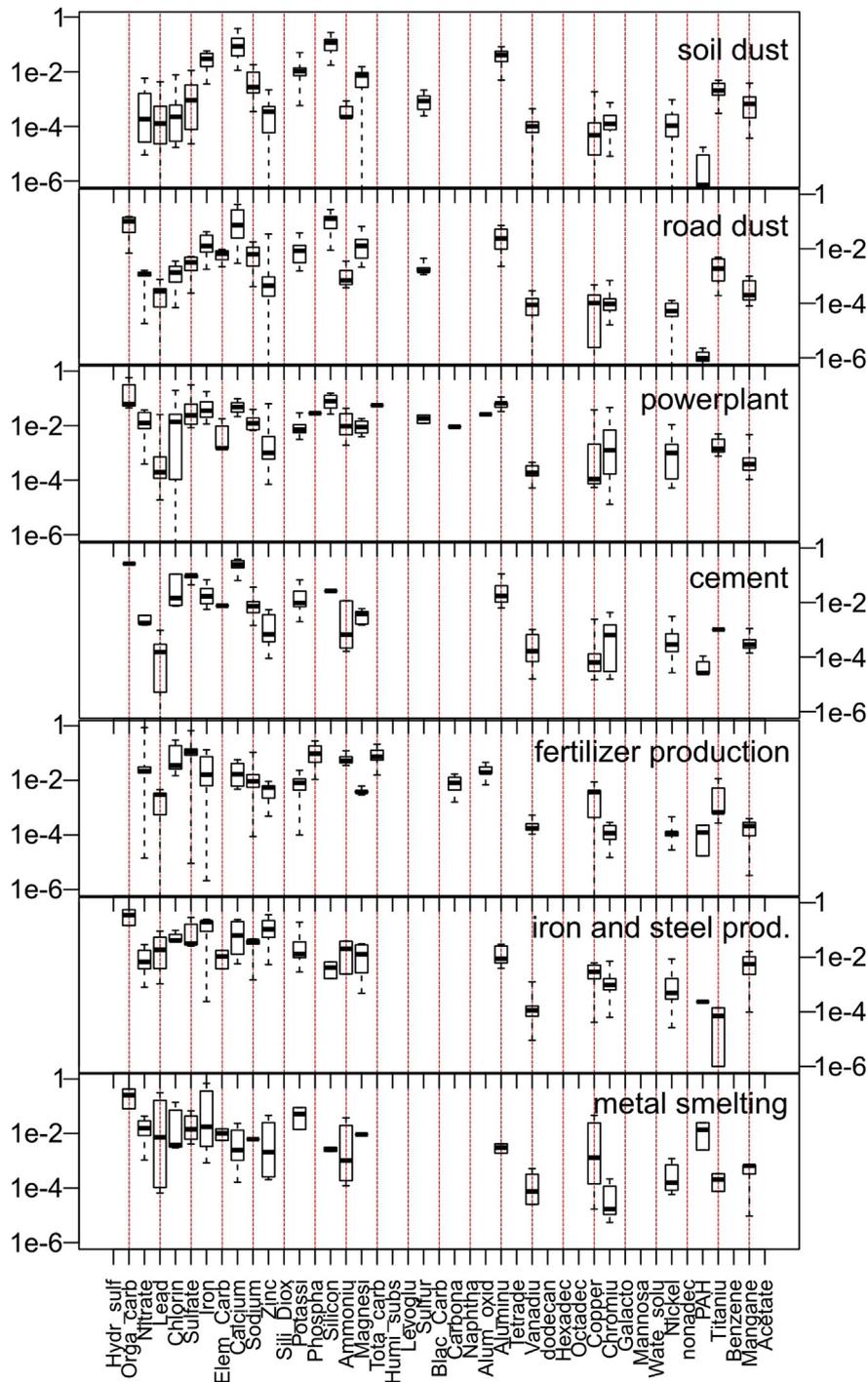


Fig. 1. Source categories associated with combustion processes. Data are in relative mass on total PM mas, logarithmic scale. Box represent median and interquartile range, whisker represent minimum and maximum values.



**Fig. 2.** Source categories associated with dust and industrial production. Data are in relative mass on total PM mas, logarithmic scale. Box represent median and interquartile range, whisker represent minimum and maximum values.

category. Fig. 3 focuses on the comparison between source categories on the basis of the relative mass of selected species aggregated by species family. The highest relative PAH relative concentrations in PM are those observed in ship exhaust followed by coke burning and wood burning (in particular soft wood). Heavy metals are most abundant in the metallurgical activities and coke burning, while halogens (mainly chlorine) are mostly present in fertilizer production, power plants and coal burning. Anhydrosugars (mostly levoglucosan) are only measured in biomass burning and related sources. The alkaline earth metals are mostly

present in road and soil dust, and in cement production (calcium). Boiler, fuel oil and ship exhaust show the largest concentrations of non-metals (sulfur).

### 3. Source categories cluster analysis

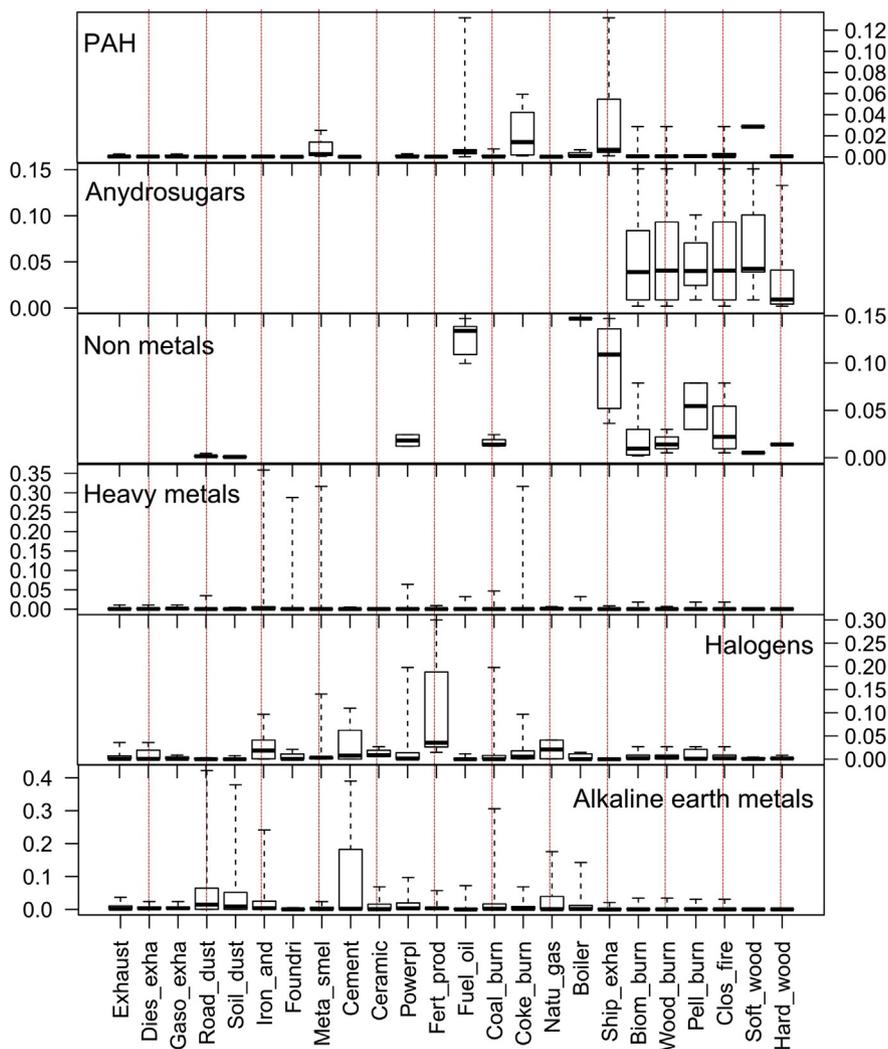
In the previous paragraph the source categories in SPECIEUROPE have been defined on the basis of the profiles' descriptions given in the original publication. In order to describe and compare the homogeneity within the various source categories the coefficient of

**Table 2**  
Species used in plots of source profiles fingerprints (first 43 up to a maximum relative mass >0.008) by IDN, name and symbol/short name.

IDN	Name	Symbol	Id	Name	Symbol
2687	Hydr. sulfate		2798	PAH	PAH
626	Organic carbon	OC	2802	Black carbon	NA
613	Nitrate	NO <sub>3</sub> <sup>-</sup>	788	Carbonate	CO <sub>3</sub> =
520	Lead	Pb	2796	Al oxide	Al <sub>2</sub> O <sub>3</sub>
337_795	Chlorine	Cl	292	Aluminum	Al
699	Sulfate	SO <sub>4</sub> =	1051	Tetradecane	N_TETD
488	Iron	Fe	767	Vanadium	V
797	Carbon	EC	599	dodecane	N_DODE
2303_329	Calcium	Ca	1045	Hexadecane	N_HEXD
785_696	Sodium	Na	1048	Octadecane	N_OCTD
778	Zinc	Zn	380	Copper	Cu
2795	Silicon Dioxide	O <sub>2</sub> Si	347	Chromium	Cr
2302_669	Potassium	K	2749	Galactosan	C6H10O5
665	Phosphate	PO <sub>4</sub>	2750	Mannosan	C6H10O5
694	Silicon	Si	2790	Water sol. OC	WSOC
784	Ammonium	NH <sub>4</sub> <sup>+</sup>	612	Nickel	Ni
2772_525	Magnesium	Mg	1047	nonadecane	N_NOND
436	Total carbon	TC	715	Titanium	Ti
2791	Humic-like	HULIS	302	Benzene	BENZE
955	Levoglucosan	levg	526	Manganese	Mn
700	Sulfur	S	3005	Acetate	

variation (CV, the standard deviation divided by the mean) was calculated for each species within the source category and the aggregated boxplots are shown in Fig. 4. The CV for the source categories represented in SPECIEUROPE go up to 5. Values above 3 are observed in industry, traffic, soil and road dust suggesting that these source categories include sources with heterogeneous chemical profiles.

Cluster analysis was applied with the aim of defining sets of profiles with homogeneous chemical composition. For that purpose, the package R pvclust (Suzuki and Shimodaira, 2006), which performs hierarchic clustering and assigns to each cluster an approximated unbiased (AU) p-value (Shimodaira, 2002) was used. The significance test is accomplished by resampling the data via bootstrap. In order to calculate the distance among profiles it is necessary that all of them have at least two species in common. For this reason, the profiles in this analysis decrease to 155. In this application of R pvclust the agglomerative method is the average, the distance is the Canberra distance (Lance and Williams, 1966; R: Distance Matrix Computation, 1998) and the bootstrap is made over 10,000 replications. The Canberra distance (CD) was selected because it is available in the R package pvclust and it is proportional



**Fig. 3.** Boxplots of the species families per source category.

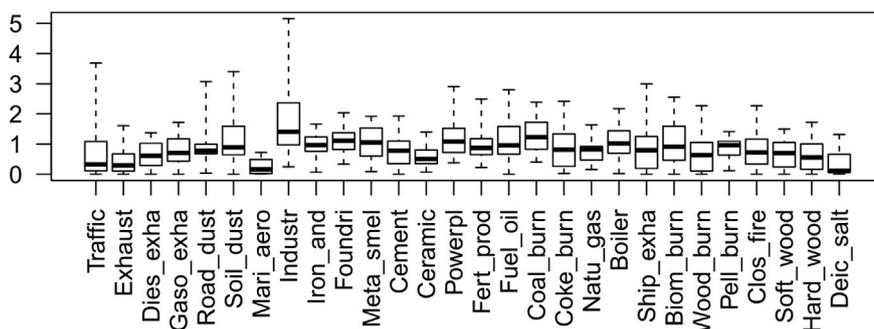


Fig. 4. Statistical distribution of the coefficient of variation for all the species in every source category.

to the standardized identity distance (SID) described in Belis et al. (2015b). The SID is defined as the ratio of the identity distance (ID) to the maximum accepted distance (MAD):

$$SID = \frac{1}{m} \sum_j \frac{ID_j}{MAD_j} = \frac{1}{m} \sum_j \frac{\frac{1}{\sqrt{2}} |x_j - y_j|}{k \frac{1}{2} (x_j + y_j)} \quad (1)$$

where  $j$  is the index of the  $m$  species in the two source profiles, and  $x_j$  and  $y_j$  are their relative masses. The  $ID_j$  measures the distance of the species  $j$  in the two profiles from the identity. The  $MAD_j$  is a criterion for the distance between two given species, defined by the user as a fraction ( $0 < k \leq 1$ ) of the average of the two species relative mass. The CD is proportional to the SID, as follows:

$$CD \equiv \sum_{j=1}^m \frac{|x_j - y_j|}{|x_j + y_j|} = \frac{km}{\sqrt{2}} SID \quad (2)$$

The SID is more suitable for comparing chemical source profiles than other commonly used indicators (e.g. Pearson correlation coefficient) because it avoids over-weighting chemical species with high concentrations. A more detailed discussion about the index is given in Belis et al. (2015b).

In Fig. 5 the sources profiles labeled with their IDN, are reported. Additional information on each cluster population

profiles is given in the Supporting Material. The clusters with distances close to zero are often profiles of different PM size fractions coming from the same publication. In a few cases, profiles from different publications present distances close zero: 4 and 200 (salt and sea water), 14 and 35 (boiler fuel oil fired), and 1 and 32 (cement kiln coal fired) because they are couples of original profiles and their composite. To avoid double counting of profiles, in the following, only one size fraction is considered and profiles with distances near to zero are considered only once. Some clusters are highlighted in Fig. 5 according to the following criteria: AU p-value  $\geq 90$  (reported in red) and profiles from at least two publications (#pub in Table 1) present in every cluster.

In order to point out the closest related species, and therefore, identify potential markers for every cluster, an analysis on the distances of each profile in the cluster from the cluster mean was performed for every species (Fig. 6). Only species available over more than 50% of profiles within the cluster were considered.

The cluster #1 (green) is identified as wood burning, especially when the sub-cluster with p-value = 96% is considered. It is composed of 13 profiles, 2 of which do not refer directly to wood burning: profile 23 is open burning of grape vine branches and profile 26 is lead smelter (coke burning). The closest species (Fig. 6) in this cluster are organic carbon (OC), levoglucosan (levg), Mg, EC, Ti and  $NH_4^+$ .

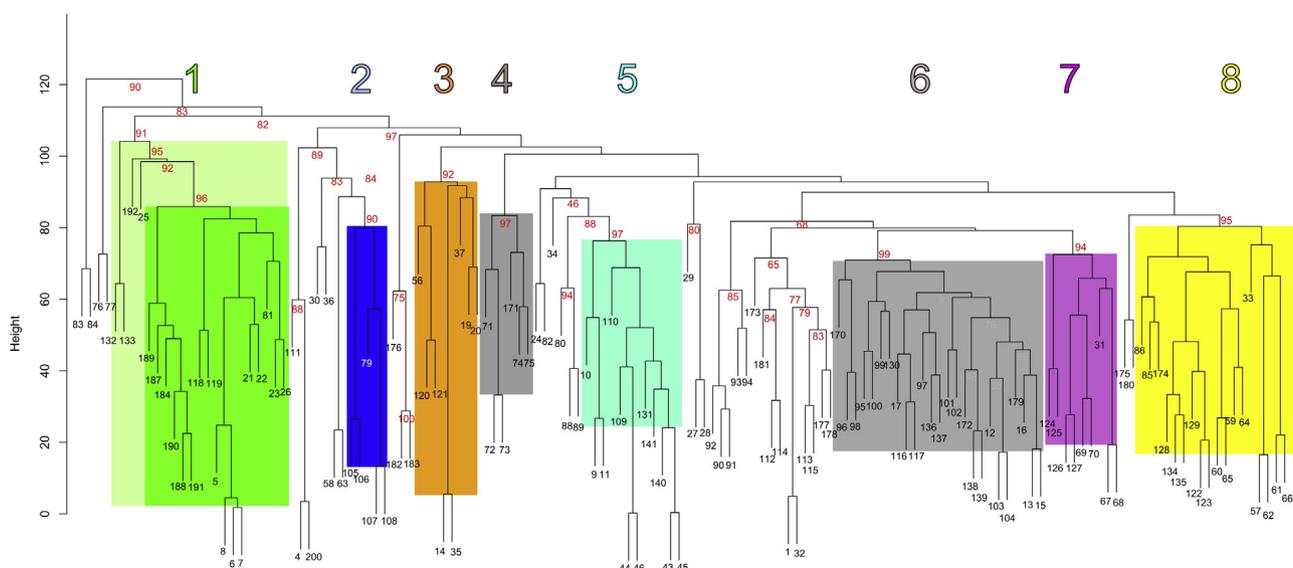
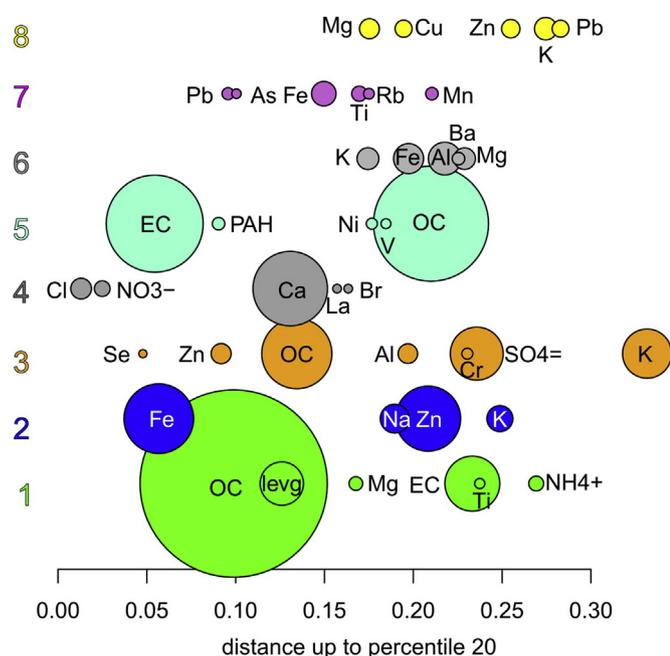


Fig. 5. Hierarchical cluster bootstrap (10,000 replications) analysis with 'canberra' distance and method 'average' on 155 profiles AU p-values are reported in red as %.



**Fig. 6.** Species with lowest distances in each cluster (numbers on the left). Balloons' radii are proportional to relative mass in logarithmic scale. Refer to Table 2 for species complete names.

The cluster #2 (blue) with a p-value of 90% is composed of only 3 profiles, all referred to steel activities. The closer species (Fig. 6) here are Fe, Na, Zn and K.

The cluster #3 (orange) is more heterogeneous than the previous two, with a p-value of 92%. The 5 included profiles are all related to combustion of different type of fuels (fuel oil, olive oil, natural gas, pellets) in a closed fireplace or boiler. The closer species in this case (Fig. 6) are Se, Zn, OC, Al, Cr,  $\text{SO}_4=$  and K.

The cluster 4 (dark grey) (p-value = 97%) contains 4 profiles (3 from a single publication) of cement and asphalt production plus a profile of urban road dust. The closer species (Fig. 6) here are  $\text{Cl}$ ,  $\text{NO}_3^-$ , Ca, La and Br.

The cluster #5 (light blue) is well defined (p-value = 97%) and is mainly traffic exhaust. It consists of 8 profiles, 2 of which are not directly related to exhaust: Aluminum rim production (109 in  $\text{PM}_{10}$  and 110 in  $\text{PM}_{2.5}$ ) and cement mill. The latter refers to the  $\text{PM}_{2.5}$  size fraction, while the companion  $\text{PM}_{10}$  (130) clusters differently. This result is coherent with the observation of the authors of these profiles (Yatkin and Bayram, 2008) who claim the characteristics of the sources are retained only in the coarse size. The closest species (Fig. 6) in this case are elemental carbon (EC), PAH, Ni, V, and OC.

Between the clusters #5 and #6 there are a number of profiles that do not significantly belong to any cluster. They mainly belong to the source categories soil and road dust (see Supporting Material).

The cluster #6 (grey) is very well defined in terms of distance (p-value 99%) and is the most populated (19 profiles): 10 soil dust profiles, 3 lignite ash and combustion profiles, 3 paved road dust profiles, a cement mill profile, a profile of dust from ceramic production and one from tobacco processing. The nature of this cluster is dominated by soil dust components. In this case the closest species (Fig. 6) are K, Fe, Al, Si, Ba, and Mg.

The cluster #7 (fuchsia) has a p-value of 94% and includes 5 profiles: 3 coal and/or coke burning profiles, one wood burning and one to fuel oil burning. The closest species in this case are (Fig. 6) Pb, As, Fe, Ti, Rb and Mn.

The cluster #8 (yellow) has a p-value of 95% and is composed of 10 profiles. Eight profiles are directly related to industrial processes: 3 related to phosphate derivatives, 2 related to coal burning, natural gas burning, oil burning and one for petrochemical and  $\text{TiO}_2$  production. Moreover, in the same cluster there are: a volcanic dust and an exhaust gasoline leaded catalytic car profiles. The closest species in this case are Mg, Cu, Zn, K and Pb.

Summarizing, clusters #1 and #5 were identified as wood burning and traffic exhaust respectively. Clusters #8, #7, #3, and #2 represent mainly industrial processes. Cement production and asphalt production are arranged in cluster #4 while cluster #6 features soil dust and road dust profiles.

#### 4. Conclusions

The freely accessible SPECIEUROPE repository is a significant step forward in the direction of giving reference chemical composition of the PM sources for source apportionment applications in Europe. More measurements to characterize sources from the chemical point of view are needed to increase the database coverage in terms of both: source categories and geographical areas. For that reasons, an effort of the scientific community to contribute to its development with existing or new source profiles is needed.

The cluster analysis confirmed the findings two intercomparison exercises (Belis et al., 2015a): a) the industry source category is quite heterogeneous and b) road dust is by definition partly composed by soil dust and therefore these two source categories are difficult to distinguish. The use of more specific markers to discriminate similar sources should be further explored.

The relevance of this database resides on its potential applications in the field of source apportionment by providing real-world reference profiles to be used either as model input or for model output validation. In addition, the availability of a common reference dataset for all the European studies is expected to contribute to a better definition of the sources and therefore to improve the comparability of studies carried out in different areas. The database finds application also in the field of emission inventories where it can be used for the validation of emission estimated derived from bottom-up approaches.

#### Conflict of interest

There is no conflict of interest.

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#### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.apr.2015.10.007>.

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