



# Indexes of tobacco smoke contribution to environmental particulates based on molecular fingerprints of alkanes

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## Abstract

Tobacco smoke (TS) is the source of a number of toxicants affecting the atmosphere and poses a threat to smokers and the whole community. Chemical, physical, and toxicological features of smoking products (vapors as well as mainstream, side stream, and third-hand smoke) have been investigated extensively. Special attention is paid to organic compounds (individually or in combination giving rise to peculiar molecular fingerprints), potentially able to act as “chemical signature” of TS. In this regard, the percent distribution of long-chain *normal*, *iso*, and *anteiso* alkanes was ascertained as typical of TS. Nevertheless, until now no indexes have been identified as suitable for assessing the global TS contribution to environmental pollution, e.g., the TS percentage in carbonaceous aerosol and in deposited dusts, the only exception consisting in the use of nicotelline as tracer. This paper describes the results of an extensive study aimed at chemically characterizing the nonpolar lipid fraction associated to suspended particulates (PMs) and deposition dusts (DDs) collected at indoor and outdoor locations. Based on the *iso*, *anteiso*, and *normal* C<sub>29</sub>–C<sub>34</sub> alkane profile in the samples as well in tobacco smoke- and no-TS-related emissions (literature data), various parameters describing the distribution of compounds were investigated. Finally, a cumulative variable was identified as the tobacco smoke impact index (TS%) suitable for estimating the TS percentage occurring in the particulate matter. The TS% rates were plotted vs. the exceedance of normal C<sub>31</sub> alkane with respect to the average of C<sub>29</sub> and C<sub>33</sub> homologs, which results higher in TS than in most other emissions, revealing a link in the case of suspended particulates but not of deposited dusts. According to back analysis carried out on all particulate matter sets, it was found that traces of TS affect even remote areas, while inside the smokers’ homes the contributions of TS to PM could account for up to ~61% and ~10%, respectively, in PM and DD. This confirms the need of valuing the health risk posed by TS to humans, by means of tools easy to apply in extensive investigations.

**Keywords** Tobacco smoke · Anteisoalkanes · Chemical fingerprints · Pollution source markers · Suspended particulates (PM) · Deposited dust (DD) · Toxicants

## Introduction

The negative health impact of tobacco smoking on the population has been ascertained, and this practice poses a threat not only to active smokers, but also to people exposed to sidestream and third-hand exhausts (IARC 1986; NCR 1986; Hecht 1999). Hence, physical and chemical properties of tobacco exhausts

have been extensively investigated (Lofroth et al. 1989; Jenkins et al. 2000), and their relationships with the toxicity of smoke have been elucidated (Husgafvel-Pursiainen 2004; Adam et al. 2006; Slezakova et al. 2009; Li et al. 2017; Pelkonen et al. 2019; Li et al. 2020; Carreras et al. 2021).

Many efforts have been dedicated to identify possible tobacco smoke (TS) tracers and know the respective relationships with cultivar, cigarette preparation, breathing time profile, etc. Chemically, TS displays a complex composition. Therefore, the first investigations were focused on TS macro-components (e.g., organic and elemental carbon, NO<sub>x</sub>, volatile hydrocarbons, and tar) (Benner et al. 1989; Eatough et al. 1990; Nelson et al. 1997, 1998; Baek and Jenkins 2004; Bi et al. 2005; Polzin et al. 2007; Moir et al. 2008; Pandey and Kim 2010; Uchiyama et al. 2018). Hundreds of micro-components have been identified in TS, including metals, alkanes, carbonyls, polycyclic aromatic

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hydrocarbons, aza-heterocyclics, and organic acids and bases (Schmeltz and Hoffmann 1977; Eatough et al. 1989; Leaderer and Hammond 1991; Rogge et al. 1994; Gundel et al. 1995; Singer et al. 2002; Ding et al. 2006; Charles et al. 2008; Lauterbach et al. 2010; Gao et al. 2015; Whitehead et al. 2015; Edwards et al. 2017; Ishizaki and Kataoka 2019).

Most of smoked organic matter is released in the gas phase due to sufficient vapor pressure of compounds combined with high temperature of fumes. However, semi-volatile substances tend to condensate as soon as the stream diffuses into the air, contributing to the organic fraction of particulate matter. The chemical toxicity of smoke components adds up to the harmful properties of airborne particulate, which is overall comprised of ultrafine particles. The organic fraction was estimated to account for up to 90% of tar and mainstream smoke, and ca. 13.5% of total TS matter (Nelson et al. 1997, 1998; Baek and Jenkins 2004; Moir et al. 2008). Among organic components, many individual substances or small groups have been proposed as tentative TS markers (Eatough et al. 1989, 1990; Daisey 1999; Douce et al. 2001; Narkowicz et al. 2012, 2013; Phillips 2012; Apelberg et al. 2013; Whitehead et al. 2015; Ishizaki and Kataoka 2019). Nevertheless, none of them looks definitively as suitable for assessing the global impact of TS on the environment. Thus, whereas it is possible to investigate the pollution in smoke chambers and TS-affected interiors through monitoring macro-components of combustion and nicotine, this approach seems unsuitable in no-smoke locations and in open air, as well as in the case of deposited dust. Indeed, these sites remain exposed to pollution sources prevailing by far vs. TS. Moreover, TS undergoes the action of light, oxidants, and water vapor altering its composition (Destailat et al. 2006; Chen and Pankow 2009; Sleiman et al. 2010; Petrick et al. 2011). A typical example of this difficulty is nicotine; this compound is volatile, and partitions between air and soot according to its concentration, ambient temperature, and chemical-physical features of particulate. Nicotine is also poorly persistent in the air, as easily attacked by acids, ozone, nitrogen oxides, free radicals, and quite soluble in water (Eatough et al. 1989; Ogden and Jenkins 1999). Despite nicotine being ubiquitous in the atmosphere and acknowledged as a tobacco marker, any attempt to link its concentrations in the air with the rate of smoking seems destined to fail.

Chemicals positively identified as typical of TS look suitable when they act as simple tracers (Benowitz 1999; Watson et al. 2004; Narkowicz et al. 2013). For instance, nicotine is accompanied by numerous alkaloids including myosmine, nornicotine, cotinine, harmanc, anabasine, anatabine, 3-ethylpyridine, and a handful of N-nitrosamines, most of which display carcinogenic potencies. Nevertheless, for quantitative purposes, a strict source of uncertainty relies in knowing the respective emission rates. Recently, a successful TS evaluation in PM has been performed by means of nicotelline (Jacob

III et al. 2013; Aquilina et al. 2021). In fact, nicotelline is enough inert, low volatile, and water insoluble to persist bound to the particle surface; besides, the emission factor of this compound is known (while its concentration ratio vs. nicotine is variable). Despite that, the loads of nicotelline in the smoke are low ( $\approx 1/1000$  of nicotine), so that this approach looks difficult to apply in extensive and routine investigations. As for polycyclic aromatic hydrocarbons, some concentration ratios (e.g., fluoranthene vs. pyrene, benz[a]anthracene vs. chrysene, benzo[a]pyrene vs. benzo[c]pyrene, and indeno[1.2.3-cd]pyrene vs. benzo[ghi]perylene) resulted enough diagnostic of TS (Ravindra et al. 2008; Sofowote et al. 2010; Tobiszewski and Namiesnik 2012; Katsoyiannis and Breivik 2014). Despite that, usually this approach cannot be applied to airborne PM and DD, because other sources of pollution are predominating in the environment (e.g., vehicles, heating and industrial plants, biomass burning), and remains potentially limited to interiors heavily affected by smoke, where other TS markers are more reliable. Solanesol is a nona-isoprenoid released by many solanaceae; however, only in tobacco plant it is important and occurs in smoke and heat-nonburnt products (Tang et al. 1990; Watson et al. 2004). Moreover, determination of solanesol content in exhausts requires dedicated procedures (ASTM 2004), which are not included in routine characterization of particulate matters.

Various authors have explored the occurrence in TS of 2-methyl (*iso*), 3-methyl (*anteiso*), and linear (*n*) long-chain alkanes, overall within the  $C_{28}$ – $C_{34}$  range, with the prevalence of odd *iso* and even *anteiso* homologs (Benner et al. 1989; Kavouras et al. 1998; Bi et al. 2005; Polzin et al. 2007; Uchiyama et al. 2018). This molecular signature accompanies the saw-tooth fingerprint of long-chain *n* alkanes [ $>C_{24}$ ], typical of natural emissions and biomass exhausts. It is known that biogenic odd-C alkanes exceed the even homologs by a factor  $\geq 5$  (Eglinton et al. 1962; Mold et al. 1964; Fine et al. 1999; McDonald et al. 2000; Oros and Simoneit 2001a, 2001b; Pio et al. 2001; Hays et al. 2002; Medeiros and Simoneit 2008; Huang et al. 2011). Besides, the TS extracts do not display the even vs. odd homologs prevalence in the short-chain range of *n* alkanes [ $\leq C_{24}$ ], typical of microorganisms (Omar et al. 2007; Bi et al. 2008; Liu et al. 2010). TS composition looks also different from those of anthropogenic emissions like fuel combustion (Gelpi et al. 1970; Reddy et al. 2000; Kuhn et al. 2010; Wang et al. 2010). Fairly indicative of tobacco smoke seems the exceedance of normal hentriacontane ( $nC_{31}$ ) with respect to  $nC_{29}$  and  $nC_{33}$  homologs. In fact, the  $nC_{31}/(nC_{29}+nC_{33})$  ratio results as high as  $\geq 1.5$  in TS, while it ranges from 0.35 to 1.15 in most anthropogenic emissions and biogenic matters.

In conclusion, very few investigations have been undertaken to estimate the TS contribution to the organic fraction of PM and DD, as well as to their whole mass. This probably depends on the insufficient availability of data concerning the composition of non-polar aliphatic fraction comprised in

emissions. Nevertheless, this knowledge would aid authorities to implement technical and legislative regulations aimed at preserving the health of citizens.

In this study, we examined several datasets of deposited dusts and airborne particulates collected both in interiors and in open air. Our focus was the nonpolar aliphatic fraction, which was characterized within the interval  $C_{24}$ – $C_{36}$ ; our goal was to explore the feasibility of drawing information about the TS impact on the environment through the alkane molecular fingerprint. In this regard, many datasets concerning the TS composition, as well as the atmospheres of the smoke chamber and internal locations, were downloaded from scientific literature and examined to identify the chemical profiles of TS and verify their specificity. The composition of the alkane fraction in TS was compared with that observed in airborne particulate samples collected inside a frequent smoke home in Roma. Various possible parameters were checked to describe the alkane fingerprint of TS, and the respective values were calculated in all sets of environmental samples analyzed. The parameters that best fitted among themselves were used to define a cumulative tobacco smoke index (TS%). Taking into account the percentages of total alkane loads in PM and DD samples, it was possible to estimate the contribution of TS to the respective particle mass. Finally, the TS% rates were plotted vs.  $C_{31}/(C_{29}+C_{33})$  ratio, to verify the occurrence of links between the two markers.

## Materials and methods

As for our airborne particulate and deposited dust series, they were collected in the frame of several investigations carried out in Italy and abroad and aimed at characterizing the respective nonpolar organic fractions. To achieve a sufficient overview of chemical profiles of alkanes associated to particulate matters, we examined very different samples with regard to the location (urban, suburban, and remote; indoor and outdoor) and the year season. The sampling procedures and analytical methods adopted to characterize the composition of airborne particulates and settled dusts are described elsewhere (Yassaa et al. 2001; Ladji et al. 2009a, 2009b; Balducci et al. 2014; Romagnoli et al. 2014; Khedidji et al. 2017). These procedures presented some differences overall with regard to the sample collection, depending on the primary purposes of the respective research projects. Briefly, airborne particulates were collected by aspirating air at low flow rates ( $2.3 \text{ m}^3/\text{min}$ ) and filtering the  $PM_{10}$  fraction onto membranes in quartz or Teflon. Deposited dusts were recovered from free surfaces according to methods developed for settled matter; free and flat surfaces ( $0.18 \div 0.50 \text{ m}^2$ ) sited at  $1.5 \div 2.0 \text{ m}$  above ground were prewashed with HPLC purity grade water, wiped with clean cotton swabs, and covered with aluminum foils previously treated with dichloromethane and acetone. Deposited

dust was recovered after 15 days of exposure to air. All samples were individually sealed in plastic bags and stored at low temperature ( $-16^\circ\text{C}$ ) until analysis. Chemical analyses consisted of the extraction with dichloromethane and acetone (80:20 in volume mixture), operated in an ultrasonic bath, separation of compounds based on molecular polarity (three fractions were eluted in sequence through a neutral alumina chromatographic column, using isooctane, isooctane/dichloromethane [80:20 in volume] and dichloromethane/acetone [50:50 in volume]), and capillary gas chromatography coupled with mass spectrometry detection (GC-MSD). In this regard, the instrumental analysis procedure was improved in order to optimize the determination of the long-chain alkanes ( $\geq C_{25}$ ). Chemical characterization was carried out in electron impact, *scan* mode over the mass/charge range  $M/Z = 44 \div 530$ ; ion currents corresponding to  $M/Z = 85$  and  $M/Z = 99$  were examined for quantitative purposes, using perdeuterated  $nC_{24}$  and  $nC_{30}$  as internal standards (using  $M/Z = 66$  and  $M/Z = 82$  signals, respectively, as reference and control traces).

Various difficulties were encountered to purchase native standards of all *iso*- and *anteiso*-alkanes investigated; therefore, the quantitative determinations of analytes were performed by using the corresponding linear homologs as reference. In this regard, the molecular responses were computed taking into account the mass spectra of compounds and the percentages of the  $M/Z$  ion traces in the respective fragmentation patterns, as they resulted from chromatograms of PM samples collected in the frequent smoker home, which resulted heavily TS contaminated. The final features of the analytical procedure are reported in Supplementary Table ST1.

Six tentative tobacco smoke parameters were defined; they are:

$$TSA_1 = \text{average}(A, B, C, D, E, F) \quad (1)$$

$$TSA_2 = \text{average}(G^{-1}, H, I^{-1}, J, K^{-1}, L) \quad (2)$$

$$\text{inCA} = \text{average}(A, C, E, S, T, U) \quad (3)$$

$$\text{anCA} = \text{average}(B, D, F, P, Q, R) \quad (4)$$

$$\text{aiCA} = \text{average}(G, H, I, J, K, L) \quad (5)$$

$$\text{SaiC} = \frac{aC_{29} + aC_{30} + aC_{31} + aC_{32} + aC_{33} + aC_{34}}{iC_{29} + iC_{30} + iC_{31} + iC_{32} + iC_{33} + iC_{34}} \quad (6)$$

where  $iC_j$ ,  $aC_j$ , and  $nC_j$  mean the respective *iso*, *anteiso*, and *normal* alkane with  $j$  carbon atoms in the molecule; besides,  $iC_{29}/nC_{29} = A$ ;  $aC_{30}/nC_{30} = B$ ;  $iC_{31}/nC_{31} = C$ ;  $aC_{32}/nC_{32} = D$ ;  $iC_{33}/nC_{33} = E$ ;  $aC_{34}/nC_{34} = F$ ;  $aC_{29}/iC_{29} = G$ ;  $aC_{30}/iC_{30} = H$ ;  $aC_{31}/iC_{31} = I$ ;  $aC_{32}/iC_{32} = J$ ;  $aC_{33}/iC_{33} = K$ ;  $aC_{34}/iC_{34} = L$ ;  $iC_{29}/aC_{29} = M$ ;  $iC_{31}/aC_{31} = N$ ;  $iC_{33}/aC_{33} = O$ ;  $aC_{29}/nC_{29} = P$ ;  $aC_{31}/nC_{31} = Q$ ;  $aC_{33}/nC_{33} = R$ ;  $iC_{30}/nC_{30} = S$ ;  $iC_{32}/nC_{32} = T$ ;  $iC_{34}/nC_{34} = U$ .

Instead of analogous diagnostic concentration ratios investigated by other authors to characterize tobacco smoke, in particular  $aC_n/iC_{n-1}$ , it was preferred to compute the  $aC_n/iC_n$  series because that approach expanded the range of values and seemed to improve the molecular signature of samples.

After the values of the six tentative parameters were calculated in all PM and DD datasets, the 90th and 10th percentiles were extracted (in order to cut possible hotspots and drops) and kept as the series maxima and minima. Then, the absolute maxima ( $Max_i$ ) and minima ( $Min_i$ ) were identified among the respective values appearing in all data sets. Finally, the delta range ( $\Delta_i$ ) was calculated for each parameter as equal to the respective difference ( $\Delta_i = Max_i - Min_i$ ).

## Results

Table 1 provides the results of this approach.

To draw a quick evaluation of the reliability of this procedure, the maxima of the above parameters were calculated by analyzing the chemical composition of tobacco smoke according to data provided by scientific literature. The corresponding values resulted in general agreement, except for the SaiC parameter, as shown in Table 2. The reasons for the SaiC behavior remain unclear; anyway, even in the literature the SaiC values were highly variable; probably, the very limited database and differences in the analytical approaches followed by the various authors partly concur to justify the difference observed.

For the sake of completeness, the values reached by each of six above parameters were calculated also in the leaves of tobacco and other plants, which was possible thanks to concentration datasets provided by the scientific literature (Eglington et al. 1962; Mold et al. 1964; Huang et al. 2011). Based on the limited data sets available, only three tentative parameters could be examined, i.e.,  $inCA$ ,  $anCA$ , and  $SaiC$ . The results, shown in Supplementary Table ST2, confirmed the specificity of the tobacco smoke chemical profile.

At this point, all samples belonging to our PM and DD data series were back analyzed, as follows: the rates reached by each  $i$ -parameter were subtracted of the respective  $Min_i$  and calculated as percentages with respect to  $\Delta_i$ , in order to obtain the corresponding first estimates of TS contribution to particulate matter. The values reached by parameters in all data sets were compared among themselves, and those resulting poorly linked were rejected, as suspected to be insufficiently specific of tobacco smoke (presumably, other emission sources released nonpolar hydrocarbons with roughly similar profiles).

Three parameters resulted finely correlated among themselves; they were  $NTSA_1 = TSA_1 / \Delta_{TSA_1}$ ,  $NTSA_2 = TSA_2 / \Delta_{TSA_2}$ , and  $NanCA = anCA / \Delta_{anCA}$ . Hence, a cumulative

**Table 1** Minimum and maximum rates (calculated as 90% and 10% percentiles, respectively) of tentative TS parameters in six environmental data series. *NoS*, number of samples

Series No.	NoS		TSA <sub>1</sub>	TSA <sub>2</sub>	inCA	anCA	aiCA	SaiC
A	98	Max	0.37	4.18	0.22	0.36	1.91	3.61
		Min	0.11	1.85	0.10	0.10	0.97	1.66
B	48	Max	0.15	4.73	0.07	0.18	1.99	4.42
		Min	0.03	2.86	0.04	0.08	1.21	1.74
C	42	Max	1.39	10.49	0.29	1.32	6.38	10.28
		Min	0.27	2.97	0.13	0.25	1.67	2.91
D	9	Max	0.30	3.17	0.14	0.32	2.77	1.10
		Min	0.11	1.83	0.08	0.13	1.95	0.95
E	13	Max	0.68	5.99	0.27	0.72	2.91	0.91
		Min	0.36	3.31	0.20	0.33	1.29	0.75
F	16	Max	0.40	5.06	0.30	0.43	3.10	4.95
		Min	0.22	2.16	0.08	0.17	0.91	1.49
G	105	Max	0.21	3.17	0.12	0.23	1.67	1.32
		Min	0.04	1.12	0.03	0.03	0.63	0.55
H	21	Max	0.42	5.32	0.23	2.62	4.74	0.28
		Min	0.16	1.79	0.10	1.29	1.74	0.09
I	4	Max	0.14	1.79	0.15	0.12	1.05	1.40
		Min	0.10	1.10	0.10	0.10	0.76	1.03
J	5	Max	0.09	1.76	0.10	0.07	0.73	0.67
		Min	0.04	0.91	0.05	0.03	0.40	0.42
K	7	Max	0.14	2.35	0.18	0.11	0.82	1.65
		Min	0.03	0.64	0.05	0.03	0.60	0.66
Total	364	Max <sub>i</sub>	1.39	10.49	0.30	1.32	6.38	10.28
		Min <sub>i</sub>	0.03	0.64	0.03	0.03	0.61	0.42
		$\Delta_i$	1.36	9.85	0.27	1.29	5.77	9.86

Rem.: (A) PM<sub>10</sub>, outdoors, Italian cities (from regional air pollution monitoring networks)

(B) PM<sub>10</sub>, outdoors, urban, suburban and rural sites from Lazio region, Italy (ARPA Lazio network)

(C) PM<sub>2.5</sub>, indoors, smoker's home, Rome, Italy

(D) PM<sub>10</sub>, outdoors, offshore sites (Mediterranean Sea)

(E) PM<sub>2.5</sub>, indoors/outdoors, hospital, Rome, Italy

(F) Dust, indoors, smoker's house, Rome, Italy

(G) Dust, indoors (homes, universities), Italian cities

(H) PM<sub>2.5</sub>, school, indoors/outdoors, Rome, Italy

(I) PM<sub>10</sub>, indoors (factory), Algiers, Algeria

(J) Dust, indoors (hospital), Rome, Italy

(K) Dust, indoors (homes, university, factory)), Algiers, Algeria

index (TSI) was defined through averaging  $NTSA_1$ ,  $NTSA_2$ , and  $NanCA$ . Moreover, the variability range of net values of the three parameters calculated for each sample allowed assessing the uncertainty of TSI estimates ( $S_{TSI}$ ).

$$TSI = \text{average}(NTSA_1, NTSA_2, NanCA) \quad (7)$$

$$S_{TSI} = \text{std.dev.}(NTSA_1, NTSA_2, NanCA) \quad (8)$$

**Table 2** Comparison of Max<sub>i</sub> rates observed in this study and in scientific literature data sets

Ref.		TSA <sub>1</sub>	TSA <sub>2</sub>	inCA	anCA	aiCA	SaiC
A	Max	1.59	9.05	0.34	1.46	7.08	6.30
	Min	1.22	8.18	0.31	1.06	6.23	6.42
B	Max	1.64	9.57	0.32	1.49	7.63	1.25
	Min	1.55	8.53	0.29	1.40	6.38	1.23
C	Max	1.29		0.29	1.30		1.26
	Min	0.89		0.27	1.07		1.24
Max. ave.	Max <sub>i</sub>	1.51	9.31	0.31	1.42	7.36	2.93
This study	Max <sub>i</sub>	1.39	10.49	0.30	1.32	6.38	10.28

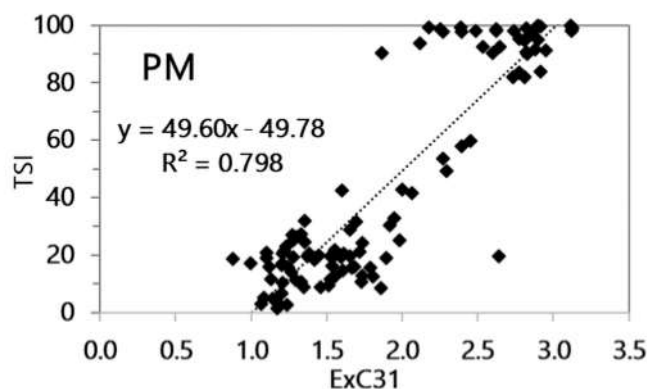
In order to check the validity of this choice, an analogous NTSI' was calculated by averaging the rates of all six parameters in each PM and DD sample; afterwards, NTSI and NTSI' values were compared. The results (summarized by Tables ST3–ST4 provided as Supplementary Material) indicate that, while the estimated rates of TS impact resulted fairly similar, the corresponding uncertainty increased by a factor of ca. 2.4 for each TSI value, while the data variability over each dataset was captured less ( $\sigma$  decreased by a factor  $\sim 0.88$ ). Thus, no improvement was reached by using all six parameters.

Chemical composition of nonpolar fraction of particulates was further characterized by means of the rate of  $nC_{31}$  homolog exceedance among the odd- $C_n$ -alkanes. For this purpose, the following formula was applied (9):

$$\text{ExC}_{31} = \frac{nC_{31}}{\text{average}(nC_{29}, nC_{33})} \quad (9)$$

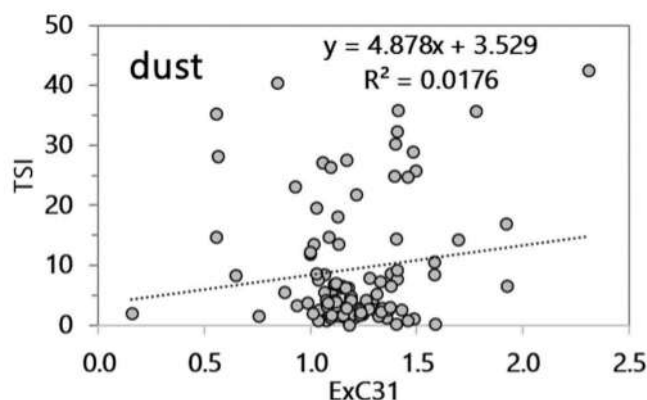
In fact, while the odd-to-even carbon prevalence looks untypical of TS exhaust but very common in biogenic emissions (e.g., in leaf debris, biomass burning, and natural waxes), the  $\text{ExC}_{31}$  index shows some specificity in tobacco smoke: we found that its rate exceeded 1.5 in tobacco smoke, while usually it was  $\leq 1$  in other biogenic and anthropogenic emissions, ranging from  $\sim 0.35$  to 1.15. Besides,  $\text{ExC}_{31}$  looked unlinked to the above parameters and independent of *iso* and *anteiso* series. The particularity of  $\text{ExC}_{31}$  rates in TS exhausts was observed by other authors; however, it remains insufficiently investigated in the context of long-chain alkanes associated to natural and anthropogenic emissions. The results of the TSI vs.  $\text{ExC}_{31}$  comparison are shown in Figs. 1 and 2, which refer to PM and dust series, respectively.

The two figures indicate that distinct relationships exist between TSI and  $\text{ExC}_{31}$  values in the cases of PM and DD. Worth to remark, the two series reported in the two figures comprised both indoor and outdoor data. Correlation plots relative to PM did not change substantially whereas the indoor and outdoor datasets were discriminated (e.g.,  $R^2$  for indoor PM raised from  $\sim 0.79$  to  $\sim 0.87$ ). Figure 1 in particular revealed the occurrence of a subgroup of PM samples, all

**Fig. 1** TSI vs.  $\text{ExC}_{31}$  correlation plot for suspended particulate matter collected at urban, suburban, and rural locations over Italy

corresponding to a frequent smoker home, for which TSI rates (all  $> 0.90$ ) lied in a pseudo plateau; on the other hand, the remaining data indicated a fair relationship between the two variables. According to that, we wanted to verify the reliability of this approach along with the whole TSI range. In fact, presumably very high TSI values corresponded to important contributions of ETS to total suspended particulate mass and vice versa. By contrast, no correlation existed between TSI and  $\text{ExC}_{31}$  in deposited dusts; this seemed in accordance with the distinct sizes of particles comprised in TS, PM, and DD matter; besides, the composition of both TS and DD is influenced by other prevailing sources, which modulate the *n*-alkane profile overall in deposited dust.

Based on literature data (Martin et al. 1997; Nelson et al. 1997, 1998; Ogden and Jenkins 1999; Jenkins et al. 2000; Back and Jenkins 2004; Moir et al. 2008), the mean contribution of the organic fraction to environmental smoke particulate matter is equal to  $13.5 \pm 0.7\%$  (Supplementary Table ST5). Meanwhile, particulate *n*-alkanes account for 21.1 mg/g (according to Li et al. 2017),  $22.5 \pm 3.2$  mg/g (Bi et al. 2005),  $28.0 \pm 8.4$  mg/g (Benner et al. 1989), and  $31.7 \pm 2.3$  mg/g (Rogge et al. 1994) (average,  $25.8 \pm 4.9$  mg/g). The literature data were in good agreement with the maximum percentage of *n*-alkanes (24.9 mg/g) associated to PM collected inside the frequent

**Fig. 2** TSI vs.  $\text{ExC}_{31}$  correlation plot for settled dusts collected at urban locations in Italy

**Table 3** Percentages of TS in total particulate mass in aerosols and dusts (TS%), tentatively calculated by assuming that *n*-alkanes account for 2.58% of total tobacco smoke matter. *PM*, suspended particulate matter (PM<sub>10</sub> or PM<sub>2.5</sub>); *DD*, deposited dust; *IN*, indoor; *OU*, outdoor; *SM*, smoking area; *NSM*, no smoking area; *SS*, spring-summer; *AW*, autumn-winter; *HO*, hospital premise; \*samplings during COVID-19 pandemic lockdown;  $\sigma$ , average TSI standard deviation in the sample set)

Series No.	Type	NoS	TS%	S <sub>TS%</sub>	$\sigma$	Remarks
A	SPM	28	0.88	0.31	0.61	OU, su., PM <sub>10</sub>
A	PM	27	0.62	0.11	0.26	OU, wi., PM <sub>10</sub>
B	PM	53	0.36	0.19	0.30	OU, sp.*, PM <sub>10</sub>
C	PM	19	22.1	2.0	9.7	IN, SM, sp., PM <sub>2.5</sub>
C	PM	8	3.50	0.37	1.69	OU, SM, sp., PM <sub>2.5</sub>
C	PM	14	60.8	5.8	14.2	IN, SM, wi., PM <sub>2.5</sub>
D	PM	14	1.22	0.24	0.53	IN, SC, PM <sub>2.5</sub>
D	PM	7	0.58	0.14	0.10	OU, SC, PM <sub>2.5</sub>
E	PM	9	1.61	0.47	0.39	IN, HO, PM <sub>2.5</sub>
E	PM	4	0.60	0.19	0.09	OU, HO, PM <sub>2.5</sub>
F	PM	8	0.44	0.11	0.14	OU, OS, PM <sub>10</sub>
G	PM	4	0.99	0.39	0.48	IN, FA, PM <sub>10</sub>
H	DD	4	10.2	1.6	3.0	IN, SH, sp.
H	DD	12	5.5	1.5	3.4	IN, SH, wi.
I	DD	57	0.26	0.13	0.44	IN
I	DD	22	0.29	0.11	0.49	OU
J	DD	5	0.08	0.05	0.04	IN, HO
K	DD	7	0.10	0.05	0.15	IN, SC

Rem.: For site identification, see Table 1

smoker home explored by us. It is worth noting that the *n*-alkane percentages resulted larger in TS than in many other emissions, in dust and PM (usually, ~0.1–5 mg/g), and in green tobacco leaves (1.25±0.75 mg/g (Severson et al. 1978)). Hence, the mass contribution of ETS to suspended particulate and dust was tentatively assessed by multiplying the TSI rates times the ratio between the net percentage of *n*-alkanes in the samples and 2.58.

The percentage of TS in PM was tentatively estimated as “percent tobacco smoke index” TS% (see Table 3).

As expected, the TS percentages in airborne particulates exceeded those associated to deposited dusts. Besides, TS percentages calculated in the interiors exceeded those found outdoors at the same sites, even in public premises (a school and a hospital). This seems to suggest that, as soon as they reach internal locations, TS components tend to persist in the environment more than in open air; meanwhile, other emissions are relatively less important there. Surprisingly, important percentages (4.4 mg/g) were found even in SPM collected during a measurement campaign performed offshore, on board a scientific ship. Finally, our results looked in general agreement with the estimates calculated by other authors for SPM (e.g., 0.1–4.0 mg/g (Aquilina et al. 2021), 10–13 mg/g (Rogge et al. 1994)), while the contribution of TS in dusts was never investigated before.

Table 3 reports the special case of frequent smoker house interiors that, based on *n*-alkane percentages in SPM (reaching a maximum of 24.9 mg/g), approached the smoke chambers.

**Table 4** Gravimetric PM concentrations and TS estimates calculated according to TS% index, particulate *n*-alkane (TNA), and nicotine (NIC) loads in a frequent smoker house

Sample No.	PM, $\mu\text{g}/\text{m}^3$	TS(TSI), $\mu\text{g}/\text{m}^3$	TS(TSI), %	TS(TNA), $\mu\text{g}/\text{m}^3$	TS(TNA), %	TS(NIC), $\mu\text{g}/\text{m}^3$	TS(NIC), %
BR1	85	28.5	33.5	32.1	37.7	35.6	41.9
BR2	105	38.3	36.5	36.7	34.9	41.3	39.3
BR3	57	19.7	34.5	23.6	41.4	10.3	18.0
BR4	74	19.5	26.3	25.1	33.9	22.7	30.6
BR5	49	20.1	40.9	17.8	36.4	9.8	19.9
BR6	115	35.7	31.1	41.0	35.7	42.1	36.6
BR7	76	37.3	49.0	32.6	42.9	8.9	11.7
LR1	101	32.2	31.9	27.1	26.8	56.6	56.0
LR2	136	41.8	30.8	38.5	28.3	66.3	48.7
LR3	67	21.3	31.8	21.9	32.7	15.8	23.6
LR4	84	17.4	20.7	24.9	29.6	31.1	37.0
LR5	56	26.0	46.5	23.9	42.6	7.2	12.9
LR6	149	50.9	36.4	60.8	40.8	78.8	52.9
LR7	78	28.4	34.2	18.4	23.6	10.5	13.5
Ave.	88.0	29.8	34.6	30.3	34.8	31.2	31.6
Std. dev.	29.9	10.0	7.4	11.4	6.1	23.3	15.3

Symbols: *BR*, bedroom; *LR*, living room; *PM*, gravimetric concentration of suspended particulate in air; *TS(TS%)*, TS concentration according to TS% rate; *TS(TNA)*, TS concentration according to total *n*-alkanes; *TS(NIC)*, TS concentration according to particulate nicotine

As expected, the contribution of TS to airborne particulate was much higher than elsewhere, though important differences were observed between the rates calculated during the spring and winter seasons ( $\sim 221 \pm 97$  mg/g vs.  $608 \pm 142$  mg/g). Besides, in the context of all the outdoor samples, the group collected at the smoker home balcony showed an important contamination coming from indoors ( $35 \pm 17$  mg/g). Due to high percentages recorded there, it was necessary to verify their reliability through using two independent approaches, based on the net concentrations of particulate nicotine and total *n*-alkanes detected in the house air during the spring season (see Table 4). As for nicotine, it was calculated that it accounts for  $\sim 2.5\%$  of total TS (Eatough et al. 1989, 1990; Rogge et al. 1994; Baek and Jenkins 2004). During this experiment, the gravimetric PM was equal to  $88 \pm 30$   $\mu\text{g}/\text{m}^3$ , and TS contribution according to TSI rate reached  $30 \pm 10$   $\mu\text{g}/\text{m}^3$  mg/g and  $346 \pm 74$  mg/g, while the fraction estimated considering nicotine and *n*-alkane concentrations was as high as  $31 \pm 23$   $\mu\text{g}/\text{m}^3$  and  $30 \pm 11$   $\mu\text{g}/\text{m}^3$ , respectively, which corresponded to  $316 \pm 153$  and  $348 \pm 61$  mg/g. When considering the overall uncertainty of the two approaches, both of them resulted in good agreement with the rated estimated on the basis of TS% rates.

The reliability of TSI/TS% approach to assess the contribution of ETS to particulate matters (to dusts in particular) merits to be further elucidated. Besides, the percentage of *n*-alkane fraction in deposited dusts and emissions needs further investigations. The typical values (maximum, minimum, and average) of ExC<sub>31</sub> index in the principal emission sources remain insufficiently known to value the use of this parameter to confirm TS pollution. Anyway, the two indexes do not depend on the same factors; both of them can be included in source apportionment studies (Kavouras et al. 2001; Mostert et al. 2010). This can be particularly useful whenever nicotine and other “classical” ETS markers presumably fail, due to volatility and degradability properties of these compounds.

## Conclusions

Six possible parameters were investigated with the purpose of assessing the contribution of ETS to airborne particulate and deposited dust in terms of the organic component and total mass. After a preliminary investigation, three of them (NTSA<sub>1</sub>, NTSA<sub>2</sub> NanCA above described) were selected to value the tobacco smoke index TS% as an estimate of ETS percentage contribution to PM and DD. Our approach seems suitable to apply in extensive in-field investigations, because it requires only the evaluation of *n*-, *iso*-, and *anteiso*-alkanes as out-of-routine chemical characterization of samples. According to the procedure above presented, airborne particulates examined were comprised of up to  $\sim 0.9\%$  of TS in open air and  $\sim 1.6\%$  in potentially smoke-free interiors; deposited dust appeared less contaminated, and ETS

accounted for up to  $\sim 0.29\%$  of the total mass in outdoor samples, and  $\sim 0.26\%$  indoors. Further investigations are needed to improve the evaluation of tobacco smoke impact on the environment through refining the values of TSA<sub>1</sub>, TSA<sub>2</sub>, anCA indexes in emissions and introducing links with other chemical signatures. Combined with ExC<sub>31</sub> and other tobacco-related parameters (e.g., nicotelline concentration in air), the TS% index seems a suitable parameter to apply in multivariate statistical approaches aimed at identifying the tobacco smoke contribution to environmental pollution.

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**Availability of data and materials** The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request. Anyway, those not directly produced by personal investigations of authors are available in the bibliography cited in the paper.

**Author contribution** This paper has been written thanks to the specific contributions of all authors. They are:

Dr. Angelo Cecinato ([angelo.cecinato@iia.cnr.it](mailto:angelo.cecinato@iia.cnr.it); [angelo.cecinato@uniroma1.it](mailto:angelo.cecinato@uniroma1.it)): result analysis, conceptualization of molecular signature indexing, original draft preparation, writing and editing; special care has been dedicated to manuscript revision

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## Declarations

**Ethical approval and consent to participate** This study did not involve experiments on humans or animals. It consisted in the analysis of literature regarding the chemical composition of tobacco smoke, as well as of data series about organic matter associated to airborne particulates and dusts, set up in the frame of research programs performed by the authors.

**Consent for publication** Not applicable. The manuscript does not contain any individual person's data, since none was subject of investigation, nor personal data treatments were performed.

**Conflict of interest** The authors declare no competing interests.

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