

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

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Abstract

Tobacco smoking is a very common habit and causes the spreading into the atmosphere of a number of toxicants, which heavily affect also health of non smokers. Hence, chemical, physical and toxicological features of smoking products (vapours as well as mainstream, sidestream and third hand smoke) have been extensively investigated. In particular, attention has been paid to organic compounds (individuals, or in combination giving rise to peculiar molecular fingerprints), potentially suitable to act as “chemical signature” of tobacco smoke (TS). At this regard, the signature of long-chain *iso* and *anteiso* alkanes has been known since long time as typical of TS. Nevertheless, until now no indexes have been identified as suitable to estimate the contribution of TS to environmental pollution, e.g. the TS percentage in carbonaceous aerosol as well as in settled dusts, nor in the respective organic fractions. This paper describes the results of an extensive investigation conducted on non-polar lipid fraction of soot and dust collected in interiors and outdoors. Six potential quantities associated to *iso*, *anteiso* and *normal* C₂₉-C₃₄ alkanes were tested, and finally a cumulative index (TSI) was defined through averaging the three most promising ones in order to estimate TS contribution to organic particulate. The behaviour of TSI was plotted vs. a further parameter, i.e. the exceedance of normal C₃₁ alkane with respect to average of C₂₉ and C₃₃ homologues, revealing a link in the case of aerosols but not of dusts. According to back analysis carried out on several sets of particulate matters, traces of TS seemed to affect even rural areas, while inside smokers' homes the contributions of TS to PM could account for up to 12.3% and 3.2%, respectively, in aerosols and dusts. This suggests the importance of further investigations aimed at assessing the potential risk for humans associated to tobacco smoking, by means of reliable TS indexes.

1. Introduction

The health impact of tobacco smoke on population is ascertained, and this practice affects not only active smokers but also humans 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 [Stedman, 1968; Lofroth et al., 1989; Jenkins et al., 2000] and their relationships with 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 tracers and know the respective relationships with tobacco cultivar, cigarette preparation, breathing time profile, etc. Tobacco smoking exhausts display a very complex composition. Therefore, investigations have focused first on macro-components (e.g., organic and elemental carbon, NO_x, volatile hydrocarbons and tar) [Johnstone and Plimmer, 1959; 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 et al., 2010; Uchiyama et al., 2018]. Among the hundreds of micro-components identified in TS, there are metals, alkanes, carbonyls, polycyclic aromatic hydrocarbons, aza-heterocyclics, 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 organic matter is released into the gas phase due to sufficient vapour pressure of compounds combined with high fumes temperature, however they tend to condensate as soon as the stream is dispersed into the air, contributing to organic fraction of particulate matter (POM). Moreover, tobacco fumes hold hundreds of semi-volatile chemicals. Chemical toxicity of smoke components adds to harmful capacity of the substrate, which is overall comprised of ultra-fine particles. POM 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 POM 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 was definitively ascertained as suitable to assess the impact of TS on the environment. Thus, though it looks quite easy to describe the pollution models of smoke chambers and interiors through monitoring macro-components of combustion or nicotine, that looks unaffordable for no-smoke locations and open air, as well as for settled dust. All of these possible environments remain exposed to pollution sources prevailing by far vs. TS. Moreover, TS experiences the action of light, oxidants and water vapour that alter its composition [Linnell, 1960; Destailat, 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. Indeed, despite nicotine has been ascertained as ubiquitous in the atmosphere and acknowledged as tobacco marker, any attempt to link its burden in the air with the rate of smoking seems destined to fail.

Chemicals positively identified as typical of TS look suitable whereas acting as simple tracers [Benowitz, 1999; Watson et al., 2004; Narkowicz et al., 2013]. For instance, nicotine is usually accompanied by numerous alkaloids including myosmine, nor nicotine, cotinine, harmaline, anabasine, anatabine, 3-ethylpyridine and a handful of N-nitrosamines, most of which of concern because of displaying carcinogenic potencies. Nevertheless, for quantitative purposes a strict source of uncertainty relies in knowing the respective emission rates from TS. For instance, an attempt has been performed with nicotelline [Jacob et al., 2013; Aquilina et al., 2021]. Though nicotelline looks as enough inert, low-volatile and water insoluble to remove from substrate, the emission factor of this compound remains uncertain and its concentration ratio vs. nicotine seems highly variable; hence, estimates of TS percentage in PM based on nicotelline content in air look suitable as indicative, but affected by wide bias.

Other chemicals seem to experience a different fate. As for polycyclic aromatic hydrocarbons, the chance of tracing TS was investigated by resorting the concentration ratios between pairs of congeners, in particular fluoranthene vs. pyrene (FA/PY), benz[a]anthracene vs. chrysene (BaA/CH), benzo[a]pyrene vs. benzo[e]pyrene (BaP/BeP), and indeno[1.2.3-cd]pyrene vs. benzo[ghi]perylene (IP/BPE). [Ravindra et al., 2008; Sofowote et al., 2010; Tobiszewski and Namiesnik, 2012; Katsoyiannis and Breivik, 2014]. Nevertheless, usually this approach cannot be applied to open air, because of the predominance of other sources of pollution (e.g., vehicles, heating and industrial plants, biomass burning), and remains limited

to interiors heavily affected by smoke. 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 burden in exhausts requires dedicated procedures [ASTM, 2004] and is out of regular routine in chemical characterization of POM.

Various Authors have underlined the presence, 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* homologues [Johnstone and Plimmer, 1959; Benner et al., 1989; Kavouras and Stratigakis, 1998; N.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 biogenic emissions and biomass exhausts. It is known that natural odd-C alkanes exceed the even ones by a factor ≥ 5 [Eglinton et al., 1962; Mold et al., 1964; Fine et al., 1999; Mc Donald et al., 2000; Oros and Simoneit, 2001 a-b, Pio et al., 2001; Hays et al., 2002; Medeiros and Simoneit, 2008; Huang et al., 2011]. TS extracts do not display the even vs. odd homologues 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 is also different from those of anthropogenic emissions like fuel combustion [Gelpe et al., 1970; Reddy et al., 2000; Kuhn et al., 2010; Wang et al., 2010]. More indicative of tobacco smoke seems the exceedance of normal hentriacontane (nC_{31}) with respect to nC_{29} and nC_{33} homologues; in fact, in most anthropogenic and other biogenic samples $C_{31}/(C_{29}+C_{33})$ ratio ranges have found ranging from 0.35 to 1.15, whilst in TS they are as high as ≥ 1.5 .

In conclusion, though physical chemical properties of TS have extensively studied, few investigations have undertaken, aimed at estimating the TS contribution to organic fraction of airborne particulates and dusts, and to their whole mass. This probably depends on the insufficient availability of data concerning the non-polar aliphatic fraction composition of particulate matter released by pollution sources. 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 sets of dusts and airborne particulates collected both in interiors and at open air, to explore the feasibility of drawing information about the TS impact on the environment. Datasets kept from scientific literature were processed with the same purpose. We have focused our attention on non-polar aliphatic fraction and characterized it within the interval $C_{24}\div C_{36}$. Various possible indexes were identified in a preliminary approach, and the respective values and patterns have assessed in the sets of samples analysed. Afterwards, a new tobacco smoke index (TSI) was calculated as the mean of rates reached by the three individual parameters resulting as sufficiently indicative of tobacco smoke emission, taking in account the values of the same parameters in the case of exhausts of various types. Besides, the TSI plots were examined through comparison with $C_{31}/(C_{29}+C_{33})$ ratio, in order to verify the occurrence of links between the two quantities in airborne particulate and settled dust.

2. Materials And Methods

The sampling procedures and analytical methods adopted by our team to characterize the composition of airborne particulates and settled dusts are described elsewhere [Balducci et al., 2014; Romagnoli et al., 2014; Kedidji et al., 2017]. Airborne particulates have collected by aspirating air at low flow rates (2.3 m³/min) and filtering the PM₁₀ size fraction onto membranes in quartz or Teflon. Dusts have recovered from free surfaces according to methods developed for settled matter; free and flat surfaces sited at 1.5-2.0 m above ground have washed in advance with HPLC purity grade water, wiped with clean cotton swabs and covered with aluminium foils previously treated with dichloromethane and acetone. The settled dust has recovered after 15 days of exposure to air. All samples have individually sealed in plastic bags and stored at low temperature (-16°C) until analysis.

Chemical analyses consisted of extraction with dichloromethane and acetone (80:20 in volume mixture), operated in ultrasonic bath, separation of compounds based on molecular polarity (three fraction were eluted in sequence through a neutral alumina chromatographic column, using isooctane, isooctane/dichloromethane and dichloromethane/acetone), and capillary gas chromatography coupled with mass spectrometry detection (GC-MSD). Instrumental analysis was carried out in *scan* mode over the mass/charge range M/Z = 44÷530; signals corresponding to M/Z = 85 and M/Z = 99 were examined for quantitative purposes, using perdeuterated *n*C₂₄ and *n*C₃₀ as internal standards. Whereas authentic *iso* and *anteiso* alkanes were unavailable, we assigned mass response factors equal to those of the respective *normal* homologues.

Six tentative tobacco smoke parameters have been defined; they are:

$$\text{TSA}_1 = \text{average}(\text{A, B, C, D, E, F}) \quad (1)$$

$$\text{TSA}_2 = \text{average}(\text{G}^{-1}, \text{H}, \text{I}^{-1}, \text{J}, \text{K}^{-1}, \text{L}) \quad (2)$$

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

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

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

$$\text{SaiC} = (\text{aC}_{29} + \text{aC}_{30} + \text{aC}_{31} + \text{aC}_{32} + \text{aC}_{33} + \text{aC}_{34}) / (\text{iC}_{29} + \text{iC}_{30} + \text{iC}_{31} + \text{iC}_{32} + \text{iC}_{33} + \text{iC}_{34}) \quad (6)$$

where: *iC_j*, *aC_j* and *nC_j* mean the respective *iso*, *anteiso* and *normal* alkane with *j* carbon atoms in the molecule; besides, *iC₂₉/nC₂₉* = A; *aC₃₀/nC₃₀* = B; *iC₃₁/nC₃₁* = C; *aC₃₂/nC₃₂* = D; *iC₃₃/nC₃₃* = E; *aC₃₄/nC₃₄* = F; *aC₂₉/iC₂₉* = G; *aC₃₀/iC₃₀* = H; *aC₃₁/iC₃₁* = I; *aC₃₂/iC₃₂* = J; *aC₃₃/iC₃₃* = K; *aC₃₄/iC₃₄* = L; *iC₂₉/aC₂₉* = M; *iC₃₁/aC₃₁* = N; *iC₃₃/aC₃₃* = O; *aC₂₉/nC₂₉* = P; *aC₃₁/nC₃₁* = Q; *aC₃₃/nC₃₃* = R; *iC₃₀/nC₃₀* = S; *iC₃₂/nC₃₂* = T; *iC₃₄/nC₃₄* = U.

Instead of analogous diagnostic concentration ratios investigated by other Authors to characterize tobacco smoke exhausts, in particular *aC_n/iC_{n-1}*, we preferred to compute the *aC_n/iC_n* series because that

approach expanded the range of values and seemed to improve molecular signature of samples.

After the values of the six tentative parameters were calculated in each data series examined, the 90th and 10th percentiles were extracted (in order to cut possible hotspots and drops), and assumed as the series maximums and minimums. Then, the absolute maximums (Max_i) and minimums (Min_i) were identified among the respective values appearing in all data series. Finally, the delta range (D_i) was calculated for each parameter as equal to respective ($\text{Max}_i - \text{Min}_i$) difference ($D_i = \text{Max}_i - \text{Min}_i$).

3. Results

Table 1 provides the results of this approach.

To obtain a quick evaluation of the reliability of this procedure, the maximums of the above parameters were calculated by analysing the chemical compositions of tobacco smoke exhausts according to data present in the literature. The corresponding values resulted in general agreement, except for *SaiC* parameter, as shown in Table 2.

The reliability of this approach was valued also by comparing the respective values in tobacco smoke, tobacco and a list of plant leaves, drawn through reprocessing literature data [Eglinton et al., 1962; Mold et al., 1964; Huang et al., 2011]. Based on the data availability, three tentative parameters were examined, i.e. *inCA*, *anCA* and *SaiC*. The results, provided in Supplementary Table ST1, confirmed the specific behaviour of tobacco smoke.

At this point, individual samples were analysed back, as follows: the rates reached by each *i*-parameter were subtracted of the respective Min_i and calculated as percentages with respect to D_i , in order to obtain the corresponding neat values. The patterns of parameters were compared each with the others, and the least linked ones have rejected, because suspected to be insufficiently specific of tobacco smoke (other emission sources presumably release non-polar hydrocarbon with roughly similar profiles). For this purpose, the one-to-one Pearson correlations among the six parameters were calculated (see Table 3).

According to them, three parameters resulted better correlated each with the other; they were TSA_1 , TSA_2 and *anCA*. Hence, the cumulative index TSI was defined equal to the average of TSA_1 , TSA_2 and *anCA*. Moreover, the range of variability of the three neat values of parameters calculated for each sample allowed assessing the uncertainty of TSI estimates (S_{TSI}).

$$\text{TSI} = \text{average}(\text{TSA}_1, \text{TSA}_2, \text{anCA}) \quad (7)$$

$$S_{\text{TSI}} = \text{std.dev.}(\text{TSA}_1, \text{TSA}_2, \text{anCA}) \quad (8)$$

In order to check for the validity of this choice, an analogous TSI' was calculated by averaging the rates of all six parameters in each particulate/dust sample; afterwards, TSI and TSI' values were compared. The results (summarized by Tables ST2-ST3 in Supplementary Material) indicate that, while the

estimated rates of TS impact were analogous, the corresponding uncertainty increased by a factor of ca. 2.4 for each TSI value associated to samples, whilst the data variability over each dataset was captured less (s decreased by a factor ~ 0.88). Thus, no improvement derived from using all six parameters.

Chemical composition of non-polar fraction of particulates has pictured also by means of the rate of nC_{31} homologue exceedance among the odd-C n -alkanes. For this purpose, the following formula has applied (9):

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

In fact, while the odd-to-even carbon prevalence is not typical of TS exhaust but very common in biogenic emissions (e.g., in leaf debris, biomass burning and natural waxes), ExC_{31} index is more specific of tobacco smoke; indeed, its rate exceeds 1.5 in tobacco smoke, while usually it is ≤ 1 in other biogenic and anthropogenic emission, ranging from ~ 0.35 to 1.15. Besides, ExC_{31} looks unlinked to the above parameters and independent of *iso* and *anteiso* series. The peculiarity of ExC_{31} rates in TS exhausts was already described elsewhere, however it still remains insufficiently matched vs. long chain profiles of n -alkanes associated to natural and anthropogenic emissions. Our results have shown in Figures 1-2, which refer to PM and dust series, respectively.

The two figures indicate that distinct relationships exist between TSI and ExC_{31} values relative to airborne particulate and dust. Worth to remark, the two series reported in the two figures comprised both indoor and outdoor data. Correlation plots relative to PM substantially did not change if indoor and outdoor series were discriminated (e.g., R^2 for indoor PM raised from ~ 0.79 to ~ 0.87). Figure 1 in particular reveals the occurrence of a subgroup of PM samples, all corresponding to a frequent smoker home, for which TSI rates (all >90) lied in a pseudo plateau; on the other hand, the remaining data indicated a fair relationship between the two quantities. According to that, we investigated the reliability of this approach along the whole TSI range. In fact, presumably very high TSI values corresponded to huge contributions of ETS to total particulate mass, and vice versa. By contrast, no correlation existed between TSI and ExC_{31} in dusts.

Based on literature data [Jenkins et al., 2000; Martin et al., 1997; Nelson et al., 1997, 1998; Ogsen et al., 1999; Baek and Jenkins, 2004; Moir et al., 2008] the mean contribution of organic fraction to environmental smoke particulate matter is equal to $13.5 \pm 0.7\%$ (Table 4). Hence, the contribution of ETS to suspended particulate and dust mass was tentatively assessed through multiplying the TSI rates times this 13.5% factor.

The percentage of TS in PM was tentatively estimated as “percent tobacco smoke index” TS% (see Table 5).

According to scientific literature, condensed phase nicotine accounts for $\sim 2.5\%$ of total PM [Eatough et al., 1989, 1990; Rogge et al., 1994; Baek and Jenkins, 2004]. Hence, whenever nicotine in the samples

exceeded 0.25% of total PM, presumably the ETS contribution to particulate matter was underestimated. This was the situation reported in Table 5, row 5, which refers to PM_{2.5} samples collected inside a frequent smoker home; they corresponded to pseudo-plateau of Figure 1. There, according to 13.5% percentage calculated above, the theoretical contribution of ETS to PM reached 12.7%, i.e. ca. 90% of organic fraction. This percentage, corresponding to >90% of 13.5 assumed as the percent rate of POM in ETS, suggests that the true percentage of ETS in PM was underestimated. To confirm this hypothesis, the particulate nicotine concentration was examined through plotting it vs. total PM measured gravimetrically (PM_{2.5} fraction). A fine correlation was found between the two quantities (see Figure 3), with a linear slope equal to 0.0094. This meant that nicotine was as high as 0.94% of tobacco smoke. Hence, according to average content of nicotine in tobacco smoke (2.5%), ETS accounted for ca. 0.94%/2.5%, namely ca. 38% of total suspended particulate and three times the percentage estimated through TS% rate. In this special case, the location seemed a smoke chamber rather than a “normal” indoor location and the TS% approach led to wrong results.

Noticeably, this situation was peculiar among the numerous samples (>300) investigated in our study. For instance, processing more than 140 PM₁₀ samples collected over Italy at outdoor locations (they were stations belonging to Regional Networks for Air Pollution Control), nicotine was as low as 0.05% of PM and never exceeded 0.16% (Figure 4); besides, no correlation was observed between the two quantities.

The range of validity of the TSI/TS% approach to assess the contribution of ETS to airborne particulates presumably needs to be further investigated. Moreover, further investigations seem still necessary to derive typical values (maximum, minimum and average) of ExC₃₁ quantity for the principal emission sources, in order to value the use of this parameter as second index of TS pollution. Anyway, despite the two indexes look as good markers of environmental contamination associated to TS, they do not depend on the same factors and may consider both in source apportionment studies [Kavouras et al., 2001; Mostert et al., 2010; Maechler et al., 2019]. This can be particularly useful whenever nicotine and other “classical” ETS markers presumably fail, due to volatility and degradability properties of these compounds. Vice versa, these two substances seem as suitable TS markers when our approach based on TSI/TS% leads to wrong results.

4. Conclusions

Six possible parameters were examined with the purpose of assessing the contribution of ETS to carbonaceous aerosol and dust in terms of organic component and total mass. After a preliminary investigation, three of them (the inCA, anCA and aiCA above described) have selected and averaged to value the tobacco smoke index TS% as an estimate of ETS percentage contribution to airborne particulates and dusts. According to the procedure here presented, airborne particulates examined were comprised of up to ~4.5% of ETS at open air and ~4.9% in potentially smoke-free interiors; dust resulted less contaminated, and ETS accounted for up to ~0.7% of total mass in outdoor samples, and ~3.2% indoors. Our approach seemed to fail in the case of frequent smoker homes, where the impact of tobacco

smoking was probably underestimated. On the other hand, nicotine and nicotelline could probably apply when our approach seemed unsuitable.

Further investigations will improve the assessment of tobacco smoke impact through refining the values of TSA₁, TSA₂, anCA quantities in emissions and introducing links with other chemical signatures. Combined with *exC₃₇* and other tobacco related parameters (e.g., nicotelline) TS% seems a promising index to apply in multivariate statistical approaches aimed at identifying the tobacco smoke contribution to environmental pollution.

Declarations

Author Contributions. This paper has written thanks to specific contributions of all Authors. They are:

Dr. Angelo Cecinato (angelo.cecinato@iia.cnr.it; angelo.cecinato@uniroma1.it): result analysis, conceptualization of molecular signature indexing, original draft preparation, writing and editing;

Prof. Alessandro Bacaloni (alessandro.bacaloni@uniroma1.it): supervision and training of doctorate thesis work;

Dr. Paola Romagnoli (romagnoli@iia.cnr.it): chemical analysis of dust and suspended particulate samples;

Tech. Mattia Perilli (perilli@iia.cnr.it): in-field campaign preparation, performance and management; paper reviewing;

Dr. Catia Balducci (balducci@iia.cnr.it): chemical procedure setup; sample analysis; investigation performance and management; writing and reviewing.

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Consent to Publish. All Authors agreed on submitting the manuscript, to publish it in ESPR Journal Editorial Board. As for external people, consent to publish was not applicable; in fact, it was not required since no experiments on humans were made nor personal data were collected or treated in the manuscript.

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.

Conflicts of Interest. The authors declare they are not under any conflict of interest condition.

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.

Ethical Approval and Consent to Participate. This study did not involve experiments on humans or animals. It consisted in analysis of literature regarding 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.

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Tables

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 ₁	TSA ₂	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	52	max	0.15	4.73	0.07	0.18	1.99	4.42
		min	0.07	2.86	0.04	0.08	1.21	1.74
c	63	max	1.27	10.49	0.29	1.32	5.31	10.28
		min	0.27	2.97	0.13	0.25	1.67	2.91
d	8	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	18	max	0.61	5.45	0.26	0.42	2.60	0.85
		min	0.05	1.50	0.06	0.03	0.63	0.56
f	15	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	0.03	1.12	0.03	0.03	0.63	0.55
<i>total</i>	363	Max _i	1.27	10.49	0.30	1.32	5.31	10.28
		Min _i	0.03	1.12	0.03	0.03	0.63	0.55
		D _i	1.24	9.37	0.27	1.29	4.68	9.73

Rem.:

- a) PM₁₀, outdoors, Italian cities (from regional air pollution monitoring networks);
- b) PM₁₀, outdoors, urban, suburban and rural sites from Lazio region, Italy (ARPA Lazio network);
- c) PM₅, indoors, smoker's home, Rome, Italy;
- d) PM₁₀, outdoors, offshore sites (Mediterranean Sea);
- e) PM₅, indoors/outdoors, hospital, Rome, Italy;
- f) dust, indoors, smoker's house, Rome, Italy
- g) dust, indoors (homes, universities), Italian cities;

Table 2. Comparison of Maxi rates observed in this study and in scientific literature data sets.

Ref.		TSA ₁	TSA ₂	inCA	anCA	aiCA	SaiC
5	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
15	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
20	max	1.32		0.29	1.30		1.26
	min	1.29		0.27	1.07		1.24
20	max	1.29			1.52		1.29
	min	0.89			1.23		1.00
21	ave.	1.14	6.72	0.32	1.00	3.91	0.99
<i>max. ave.</i>	Max _i	1.35	8.45	0.32	1.35	6.21	2.22
<i>this study</i>	Max _i	1.27	10.49	0.30	1.32	5.31	10.28

Table 3. Pearson correlation coefficients (R) among the six TS parameters, calculated on our whole data set.

R	TSA ₁	TSA ₂	inCA	anCA	aiCA	SaiC
TSA ₁	1.00	0.75	0.75	0.99	0.70	0.67
TSA ₂	0.75	1.00	0.42	0.76	0.66	0.63
inCA	0.75	0.42	1.00	0.71	0.34	0.32
anCA	0.99	0.76	0.71	1.00	0.71	0.68
aiCA	0.70	0.66	0.34	0.71	1.00	0.85
SaiC	0.66	0.63	0.32	0.68	0.85	1.00

Table 4. Mass percentage of organic fraction in tobacco smoke, according to scientific literature.

Ref. No.	%	std.dev.
6	13.1	0.6
14	12.4	0.7
15	13.6	0.6
23	14.5	0.7
24	14.3	0.9
82	13.2	0.7
83	13.4	0.4
average	13.5	0.7

Table 5. Percentages of TS to total particulate mass in aerosols and dusts (TS%), tentatively calculated by assuming that organics fraction accounts for 13.5% of tobacco smoke matter. SPM = suspended particulate matter (PM₁₀ or PM_{2.5}); 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; s = average TSI standard deviation in the sample set).

Series No.	type	NoS	TS%	S _{TSI}	s	remarks
A	SPM	49	2.36	0.44	0.87	OU, SSr
A	SPM	49	2.29	0.33	0.79	OU, AW
B	SPM	52	0.89	0.14	0.61	OU, SS*
C	SPM	34	12.7	0.4	0.7	IN, SM
C	SPM	16	4.21	0.20	2.12	OU
C	SPM	16	3.09	0.53	0.82	IN, NSM
D	SPM	8	1.90	0.25	0.89	OU, SS
E	SPM	9	4.90	1.19	1.14	IN, AW, HO
E	SPM	4	2.78	0.78	0.55	OU, AW, HO
F	dust	15	3.59	0.43	0.89	IN, SM
G	dust	41	0.92	0.42	1.09	IN, SS
G	dust	14	0.89	0.27	1.48	OU, SS
G	dust	42	0.86	0.31	0.87	IN, AW
G	dust	8	0.75	0.23	1.19	OU, AW

Rem.: For site identification, see Table 1.

Figures

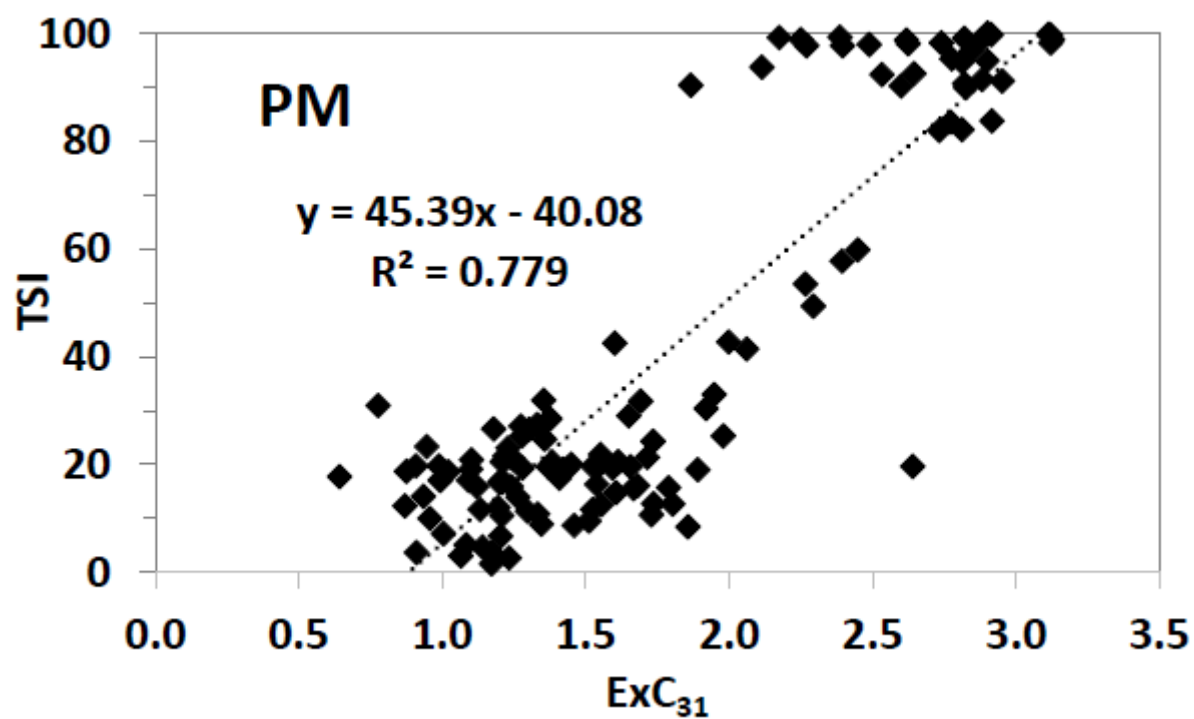


Figure 1

TSI vs. ExC31 correlation plot for suspended particulate matter collected at urban, suburban and rural locations over Italy

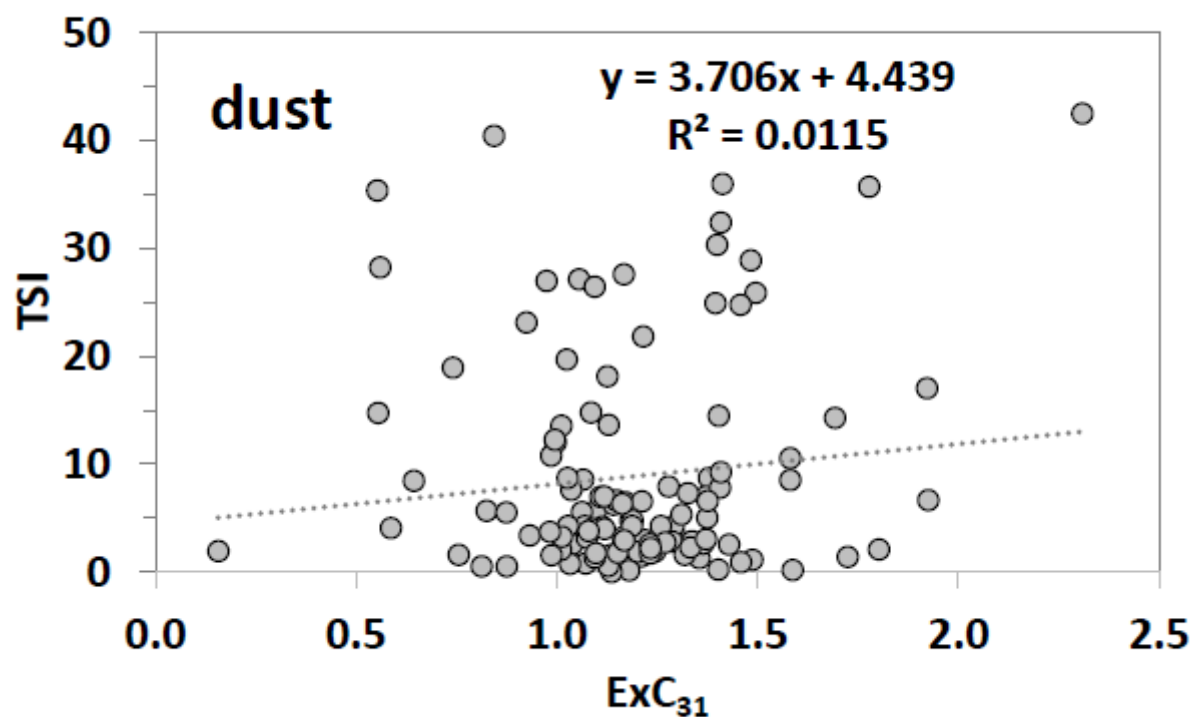


Figure 2

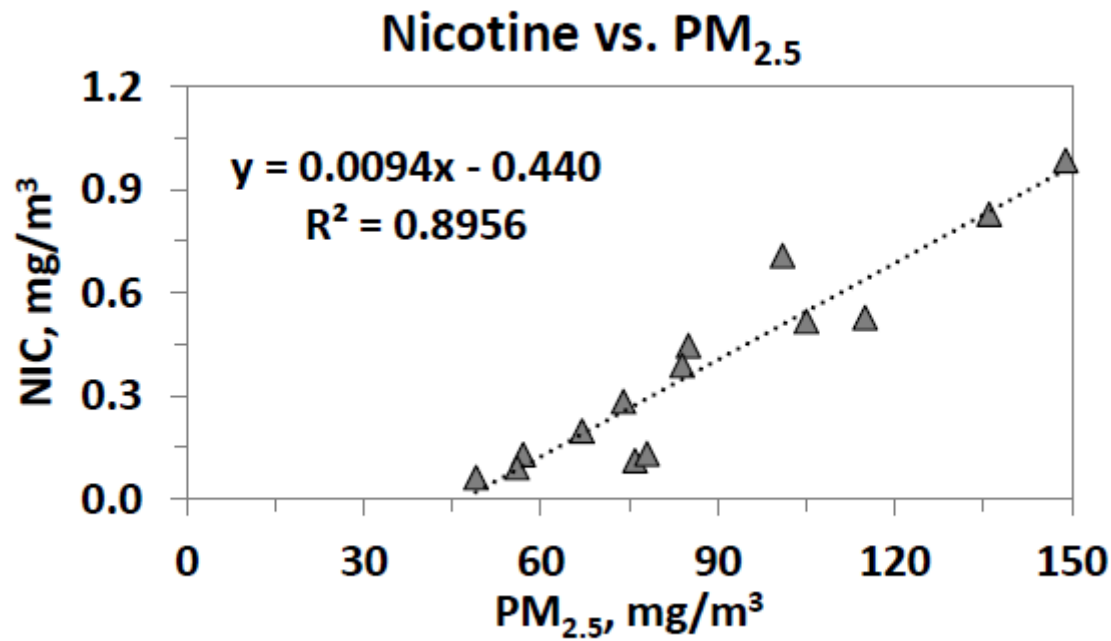


Figure 3

Nicotine vs. PM_{2.5} plot in a frequent smoker home (see row no. 5 in Table 5).

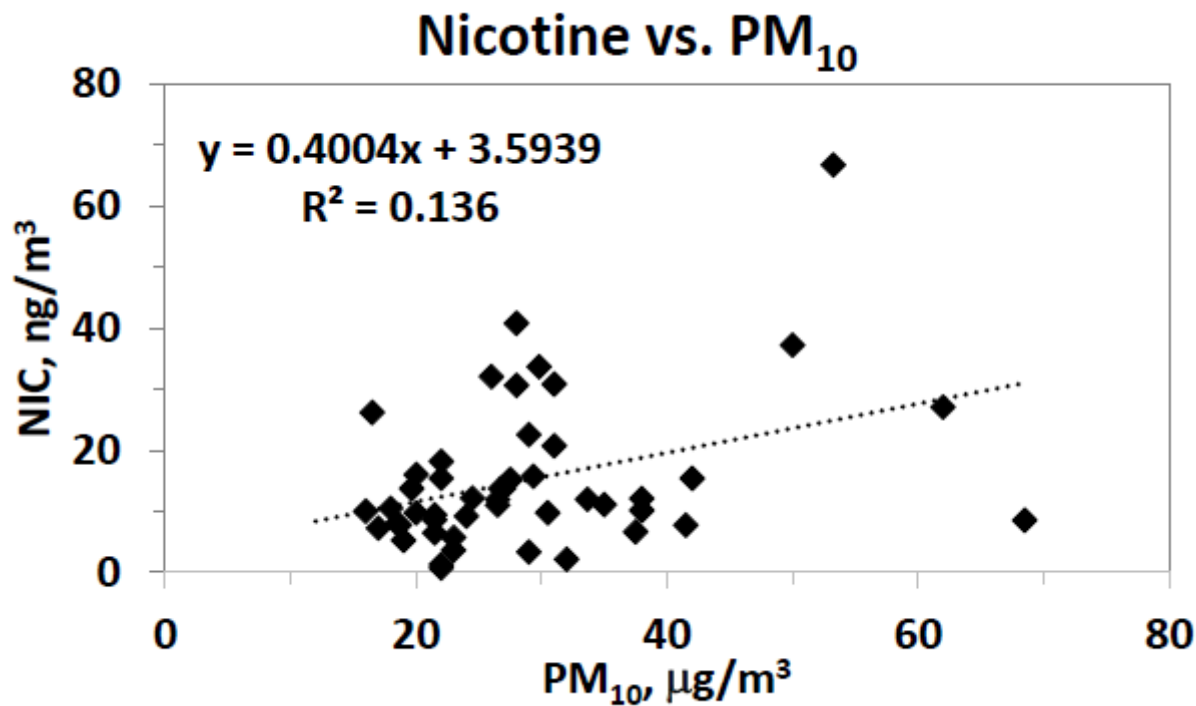


Figure 4

Nicotine vs. PM_{2.5} plot in several locations over Italy (outdoor sites).

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