

Full Length Research Paper

# Effect of the Amount of Tobacco per Cigarette on Smoke Composition when Smoking 3R4F Reference Tobacco and a Commercial Tobacco Brand with and without a Catalyst.

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## ABSTRACT

The effect of the weight of tobacco per cigarette (WTC) on the mainstream smoke of reference (R) and commercial (F) cigarettes and the effect of the addition of an SBA-15 catalyst has been studied. The yield of total particulated matter (TPM) obtained from the reference tobacco, with and without catalyst, is decreased as WTC increases, but the results for the commercial cigarettes show a maximum for TPM and CO at the medium WTC value. The highest yields of the different chemical families of compounds appearing in TPM extracted from filters (TPM-F) from F and R cigarettes, and in TPM from the traps after filters (TPM-T) from R, have been obtained at the intermediate WTC, whereas for TPM-F from reference cigarette + catalyst (RC) and TPM-T from F and RC, a decreasing trend as WTC increases is observed. Interesting reductions have been obtained in the presence of catalyst, especially for the carbonyls group in TPM-F and for aromatics in TPM-T.

**Key words:** Tobacco weight per cigarette, Catalyst, Smoke tobacco, Gaseous fraction, Particulate matter.

## INTRODUCTION

Tobacco is a very popular product that attracts great interest from many points of view. Tobacco products represent a significant share of the world economy, considering both aspects, the costs involved in advertising, promotion and tobacco use as well as the medical expenses and decrease of productivity. Tobacco also concerns public health departments, and was declared the leading cause of foreseeable death by the World Health Organization (WHO) in 1956<sup>[1]</sup>. Additionally, tobacco is one of the most controversial commercial products and is becoming the object of an increasing number of bans, restrictions and regulations in different countries<sup>[2]</sup>. In fact, it would not meet the existing regulations to become a commercial product in many countries, if nowadays it was attempted to be sold for the first time. As Purkis et al.<sup>[3]</sup> have pointed out; tobacco smoke components are associated with a large amount of terrible diseases.

Many processes take place when tobacco is smoked. Among them, pyrolysis/distillation, combustion, pyrosynthesis, condensation and dilution, are the main events responsible for the amount and composition of tobacco smoke<sup>[4, 5]</sup>.

The smoking regime or the smoking characteristics of the smokers have a noticeable impact on the amount and chemical composition of the obtained tobacco smoke<sup>[6]</sup>. Moreover, the type of tobacco leaves, blends and additives<sup>[7]</sup>, the type of paper and filters, the presence of ventilation holes in the filters<sup>[8]</sup>, and the use of additives at different levels<sup>[9]</sup> also play a very important role in the smoke yields and composition. The influence of the weight of tobacco per cigarette (WTC) has been considered in several works<sup>[8, 10]</sup>, although in these cases the role of the rod diameter and other design parameters were also considered. In general, the smoke yields increase as the cigarette tobacco content increases, despite the relationship not being linear, because of the influence of the paper porosity and the differences in rolling techniques.

Among the different methods suggested for reducing the toxicity of tobacco smoke, the use of additives as zeolites or related

materials has been considered. As an example, the effect of several zeolites and other aluminosilicates on the evolution of different compounds such as nitrosamines, CO and other toxic compounds as polyaromatics or polyphenols has been reported <sup>[11-16]</sup>. A ferric zeolite has also been proposed for the catalytic degradation of tobacco-specific nitrosamines <sup>[17]</sup>.

In previous works <sup>[18-22]</sup>, we have shown the effect of different catalysts on the yields and proportion of different compounds appearing in tobacco smoke. The results reported in these papers demonstrate that the addition of certain materials directly to the tobacco allows the reduction of most of the compounds detected in the mainstream tobacco smoke. In these studies, different catalysts were considered, and the influence of the amount of catalyst used <sup>[22]</sup>, as well the synthesis conditions have also been considered <sup>[18-20]</sup>. As has been pointed out in the previous paragraphs, it is well known that the WTC is an important parameter affecting the yields of the different compounds in the smoke as well as the total smoke amount <sup>[8, 10]</sup> because it has a significant incidence on the compacting degree and, therefore, on the diffusional and other processes affecting the mass transfer of air and pyrolysis and combustion products through the cigarette, and on the pressure drop associated with each puff. Nevertheless, to our knowledge, there are no bibliographic references studying the influence of this variable when a solid catalyst is used as a cigarette additive in order to reduce the concentration of different evolved compounds. Thus, this work is focused on the study of the effect of varying the WTC, keeping constant the other design parameters, with the only exception being the void volume inside the rod of the cigarette, that obviously decreases as WTC increases, on the yields of different compounds appearing in the mainstream smoke, and its incidence when a catalyst able to reduce the tobacco smoke toxicity is used as a cigarette additive.

## EXPERIMENTAL

### Materials

Two different types of tobacco have been used, one of them is the "full flavor", filtered, American blended reference tobacco 3R4F from the University of Kentucky <sup>[23]</sup>, and the other is one of the most popular Spanish commercial tobacco brands, that also contains a blend of American tobacco. Before performing the smoking experiments, 100-200 cigarettes of each type were disassembled, and the tobacco, the filter and the paper were weighed separately. Tobacco was tumbled and mixed and then, new cigarettes were manually reassembled with the tobacco or with mixtures of tobacco and catalyst (around 6 wt%), using three WTC levels (the average WTC for each tobacco type, the average +0.1 g/cigarette and the average -0.1 g/cigarette, i.e., cigarettes containing around 0.66, 0.76 and 0.86 g/cigarette in the case of the reference tobacco, and at around 0.60, 0.70 and 0.80 g/cigarette in the case of the commercial brand. The cigarettes were prepared using the corresponding and previously emptied tubes and had been conditioned for at least 48 h at 22°C and a relative humidity of 60%. Afterwards the cigarettes were smoked under the ISO 3308 standard conditions, and the chemical composition of the gases obtained and that of condensed products retained in the filters and in the traps was analyzed.

According with the nomenclature used in this work, R and F refer to the cigarettes prepared with the reference tobacco and with that from the commercial cigarettes. RC identifies the cigarettes prepared with the reference tobacco mixed with the catalyst. In the tables, the underlined sample corresponds to that containing a WTC value very close to that of the original reference or commercial cigarettes.

The catalyst checked is a mesoporous SBA-15 catalyst, self-synthesized according to the patent <sup>[22]</sup>, by dissolving a triblock poly (ethylene oxide)-b-poly (propyleneoxide)-b-poly (ethylene oxide) copolymer (sigma-aldrich) in water and HCl solution. After that, TEOS (Tetraethyl ortosilicate 99%, Aldrich) was added, and the mixture was maintained at 38°C, stirred for 20 h, and after at 100°C for 24 h. The white solid products were collected by filtration, dried at 100 °C, and then calcined at 550°C for 5h. The textural characteristics of this material were determined by the measurement of the N<sub>2</sub> adsorption isotherm at 77 K in an automatic Quantachrome AUTOSORB-6. The adsorption curves of the isotherms were recorded and the surface area was determined according to the BET method. The pore size distribution was obtained by applying the BJH model with cylindrical geometry of pores and using the de Boer equation for determining the adsorbed layer thickness (t) and the external surface area. Table I shows the corresponding physical properties.

**Table I:** Textural parameters of the catalyst

Pore Size (nm)	6.09
BET area (m <sup>2</sup> /g)	757
Total Pore volume (cm <sup>3</sup> /g)	1.06

### Smoking Experiments

A smoking machine has been employed <sup>[21]</sup> that allows five cigarettes to be smoked simultaneously, with a pressure of aspiration of the machine that was never higher than 1.5kPa. Fifteen cigarettes were smoked for each experiment. The puff volume was 35 mL, taken for 2.0 seconds, with a puff frequency of 60 seconds, according to the ISO 3308. The cigarettes were placed in the ports of the smoking machine ensuring that the ventilating holes were not blocked, and then the cigarettes were smoked. The

standard butt length, to which cigarettes shall be marked, must be over 23 mm. The mainstream smoke, after have passed through the cigarette filter, was passed through another trap in order to retain the less volatile compounds that could condense in the mouth and lungs of smokers, and collected in a Tedlar bag, as has been described elsewhere<sup>[21]</sup>.

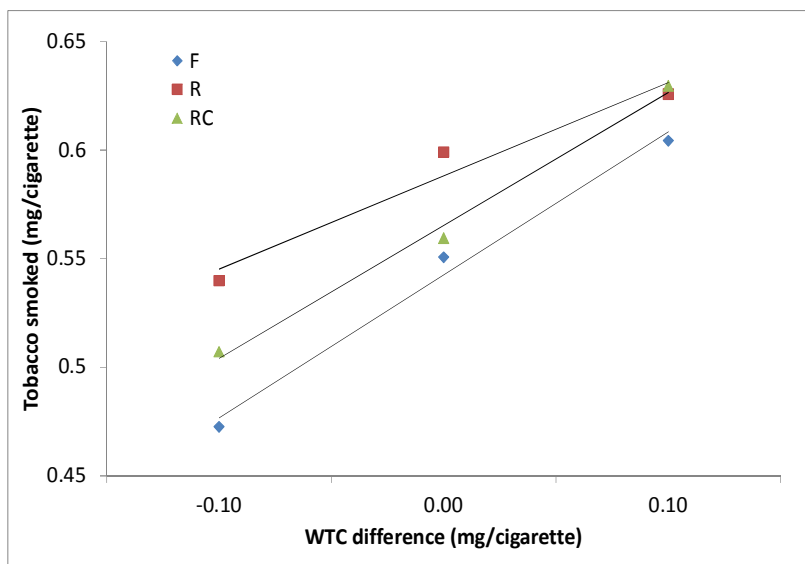
The global yields of condensed products in the filters and in the traps in each experiment are obtained by the weight difference before and after the experiment. The amount of tobacco smoked is calculated as the difference between the initial mass of tobacco in the cigarette and the tobacco remaining in the cigarette butt after each experiment. The gas collected in the Tedlar bag is analyzed by gas chromatography (GC) with two different types of detector and chromatographic columns, i.e., a thermal conductivity detector (GC/TCD) and a CTR I column in order to determine the CO and CO<sub>2</sub> yields, and a flame ionization (GC/FID) and a GAS-PRO column in order to determine other components of the gaseous fraction of the mainstream smoke. More details on the analysis conditions are reported elsewhere<sup>[21]</sup>. The measurement of the yields of nicotine and other condensed compounds, both in the filters and traps, has been performed according to the ISO 4387 that describes the procedure for the analysis of the total particulate matter (TPM), nicotine and water. TPM is defined as the fraction of the mainstream smoke of tobacco which is retained in the traps, expressed in mg/cigarette. In this work, the TPM has been extracted from filters (TPM-F) and from the traps (TPM-T) with 2-propanol as a solvent, and has been analyzed by gas chromatography-mass spectrometry (GC/MS), using a HP-5MS column. 34 and 83 compounds were identified in the fraction collected in the Tedlar bags (i.e. the gas fraction) and in the TPM (both, TPM-F and TPM-T), respectively. The analysis and quantification of the analytes have been carried out as was previously reported<sup>[21]</sup>.

## RESULTS AND DISCUSSION

Table II shows the WTC of the different cigarettes smoked, the amount of smoked tobacco, and the yields of TPM-F, TPM-T and CO obtained from the smoking experiments for the cigarettes prepared with the two types of tobacco studied in this work (R and F) and for the cigarettes prepared with the reference tobacco and the catalyst (RC). In Figure 1, the amount of smoked tobacco in front of WTC has been represented, showing the expected result that the amount of tobacco smoked increases almost linearly with WTC in all cases. Moreover, the slope corresponding to the F tobacco is higher than that of the R tobacco, and similar to the RC cigarettes, thus suggesting that, despite in all the cases the amount of tobacco smoked is lowered by the catalyst addition; it seems that the catalyst favors the increase of the smoking process as WTC increases.

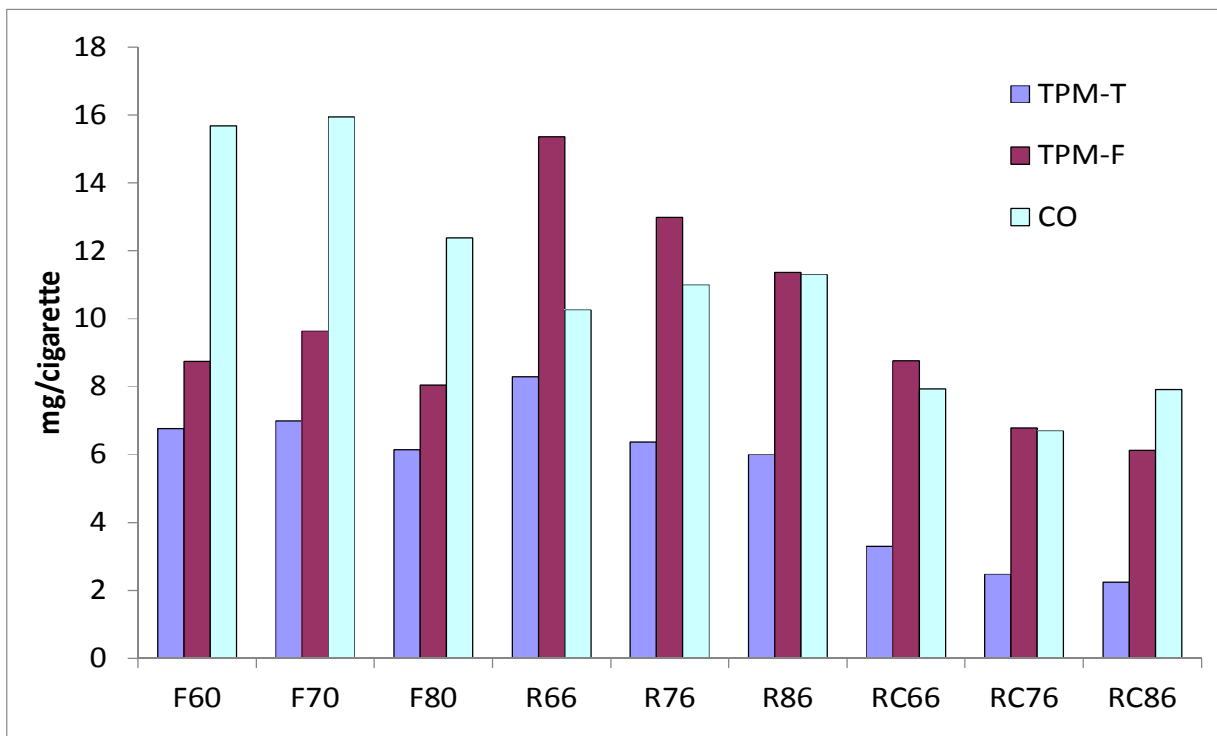
**Table II:** Weight of tobacco per cigarette (WTC), amount of smoked tobacco, and yields of TPM-F, TPM-T and CO obtained for F, R and RC cigarettes.

SAMPLE	WTC (g/cigarette)	smoked tobacco (g/cigarette)	TPM-T (mg/ cigarette)	TPM-F (mg/ cigarette)	CO (mg/ cigarette)
F60	0,604	0.472	6.77	8.75	15.69
F70	0,705	0.551	6.99	9.64	15.95
F80	0,804	0.605	6.15	8.05	12.38
R66	0,662	0.540	8.29	15.36	10.26
R76	0,767	0.599	6.38	12.98	11.00
R86	0,862	0.626	6.00	11.36	11.30
RC66	0,663	0.507	3.30	8.77	7.94
RC76	0,763	0.559	2.47	6.78	6.70
RC86	0,852	0.630	2.24	6.13	7.91



**Figure 1:** Amount of smoked tobacco versus WTC for F, R and RC. The WTC difference values of X axis represents the difference between the WTC of the smoked cigarettes and the WTC mean value in the commercial cigarettes.

Figure 2 shows a graphical comparison among the yields of TPM-T, TPM-F and CO for F, R and RC with the three values of WTC considered. It can be seen that, in all the cases, the amount of carbon monoxide obtained from F is higher than that obtained from R, whereas TPM-F is lower for F than R. The yield of TPM-T is relatively similar for both cases, and depending on the range of WTC could be higher for R (for the lowest value of WTC) or for F. The analysis of the effect of the catalyst reveals a noticeable decrease of the yields of CO, TPM-F and TPM-T when we compare the results corresponding to R and RC. This behavior is what should be expected from the previous studies<sup>[22]</sup> and enhances the ability of the catalyst for its use as an additive for reduction of tobacco toxicity. In fact, global reductions of around 60%, 40% and 30% are obtained for TPM-T, TPM-F and CO, respectively. The general trend observed for the yields of CO, TPM-F and TPM-T when WTC increases is as follows: in the case of F-cigarettes, there appears a maximum at the medium WTC-value, whereas in the case of R and RC-cigarettes, the yields of TPM-F and TPM-T decrease and the yield of CO is almost unaffected and only shows a very slight increase for R and a minimum for RC.



**Figure 2:** TPM-T, TPM-F and CO (mg/cigarette) for F, R and RC tobaccos

### Analysis of the Gaseous Fraction of the Mainstream Smoke

The analysis of the gaseous samples collected in the Tedlar bags from the different smoking tests has been performed in the conditions described in the previous sections. The results obtained are shown in Table III, expressed as mg of compound per cigarette. The total volatile organic compounds (VOC) have been evaluated as the sum of the yields of all the analyzed compounds, and have also been included in Table III. As can be seen, there is no general tendency, and as WTC increases, the VOCs from R also increase, but the VOCs from F and from RC pass through a maximum and a minimum, respectively. It is worth mentioning the noticeable decrease of the VOCs from R when the catalyst has been used as a cigarette additive, which reflects that most of the individual compounds are decreased, despite some compounds, such as iso-butane and hexane, could be increased. In order to facilitate the interpretation of the results shown in Table III, the different compounds have been grouped by chemical families, as paraffins, olefins, aromatics, aldehydes and other compounds. Figures 3 and 4 show the yields obtained for each family from the different types of cigarettes studied (i.e., F, R and RC) versus the WTC. As it can be seen, paraffins is the family showing the highest yields, being higher for R than for F and RC. The values obtained for olefins are very close for the three systems studied, and the amounts of the other families are in the order of a tenth of hydrocarbons. The trend observed when WTC increases is the same for all the groups, and increases for R, passing through a minimum for RC and through a maximum for F. In our knowledge, there are no studies in the literature focused on the influence of the weight of tobacco on the yield of the different compounds appearing in the mainstream smoke, and only the study of the effect of two different masses of RYO tobacco on the yields of CO, tar and nicotine has been found<sup>[10]</sup>. These authors reported that, as expected, cigarettes with higher tobacco content, produced higher smoke yields than those made with lower tobacco amount, regardless of the cigarette paper chosen for the comparison. However, they found that, in general, the yields did not decrease in proportion to the weight of tobacco, particularly in the case of the more porous paper, and pointed out that these findings must be interpreted cautiously because of the difference in rolling technique that was necessary for the lower weight of tobacco. Also, some association between the mean weight of tobacco per cigarette and the yields of CO, nicotine, and tar, that showed a slight upward trend in yield with increasing weight, was found, in spite of the fact that some cigarettes that were loosely packed showed yields lower than might have been predicted. These results reflect the noticeable influence of the cigarette parameters with influence on the air availability for pyrolysis and combustion processes. Thus, the differences found in the behaviour of F and C would not only be associated with the type of tobacco blend, but also with the type and permeability of paper, and enhances the difficulty of generalizing the results of this type of study and the need to include each type of tobacco or cigarettes of interest as the subject of the studies.

**Table III:** Yield of the different compounds analyzed in the gaseous fraction of the mainstream smoke from F, R and RC

COMPOUNDS (mg/cigarette)	Family	F60	F70	F80	R66	R76	R86	RC66	RC76	RC86
Methane	Paraffin	0.830	0.953	0.899	1.168	1.301	1.482	1.015	0.850	0.928
Ethane	Paraffin	0.339	0.403	0.394	0.474	0.532	0.613	0.384	0.340	0.377
Ethylene	Olefin	0.194	0.217	0.199	0.238	0.274	0.304	0.250	0.201	0.204
Acetilene	Other	0.021	0.022	0.022	0.026	0.031	0.033	0.044	0.035	0.027
Propane	Paraffin	0.156	0.188	0.187	0.213	0.239	0.276	0.172	0.151	0.167
Propene	Olefin	0.184	0.212	0.206	0.228	0.263	0.297	0.217	0.180	0.192
Iso-butane	Paraffin	0.017	0.017	0.037	0.019	0.031	0.024	0.055	0.053	0.016
Chloromethane	Other	0.042	0.048	0.049	0.061	0.065	0.073	0.051	0.042	0.046
Butane	Paraffin	0.049	0.058	0.070	0.064	0.072	0.083	0.051	0.048	0.054
1-butene	Olefin	0.045	0.052	0.051	0.054	0.063	0.072	0.049	0.040	0.044
1,2-Propadiene	Olefin	0.009	0.009	0.008	0.009	0.013	0.012	0.015	0.009	0.009
1,3-Butadiene	Olefin	0.013	0.016	0.013	0.012	0.020	0.020	0.019	0.012	0.012
Isobutene	Olefin	0.042	0.051	0.049	0.052	0.061	0.070	0.048	0.039	0.045
Cis-2-butene	Olefin	0.030	0.039	0.036	0.032	0.045	0.042	0.032	0.027	0.032
Pentane	Paraffin	0.013	0.018	0.017	0.018	0.020	0.025	0.014	0.013	0.014
Methanethiol	Other	0.014	0.015	0.010	0.016	0.017	0.017	0.026	0.013	0.014
Hydrogen cyanide	Other	0.010	0.012	0.013	0.012	0.015	0.017	0.013	0.009	0.011
1-Pentene	Olefin	0.013	0.017	0.015	0.014	0.017	0.020	0.015	0.010	0.013
Furan	Aromatic	0.015	0.021	0.017	0.019	0.022	0.029	0.018	0.010	0.016
Isoprene	Olefin	0.292	0.353	0.318	0.195	0.236	0.321	0.293	0.088	0.204

Table III: Yield of the different compounds analyzed ... (Cont.)

Hexane	Paraffin	0.013	0.016	0.015	0.006	0.007	0.011	0.013	0.008	0.006
1-Hexene	Olefin	0.013	0.015	0.014	0.012	0.017	0.016	0.012	0.010	0.011
Benzene	Aromatic	0.124	0.130	0.120	0.147	0.160	0.169	0.131	0.092	0.094
Acetaldehyde	Aldehyde	0.435	0.515	0.523	0.606	0.741	0.731	0.601	0.378	0.453
Acrolein	Aldehyde	0.026	0.029	0.026	0.032	0.053	0.037	0.068	0.052	0.049
Propionaldehyde	Aldehyde	0.038	0.041	0.035	0.038	0.035	0.048	0.035	0.024	0.023
Acetonitrile	Other	0.069	0.084	0.066	0.076	0.049	0.064	0.036	0.022	0.021
toluene	Aromatic	0.025	0.029	0.029	0.029	0.038	0.037	0.025	0.024	0.021
2,5-Dimethylfuran	Aromatic	0.012	0.015	0.013	0.012	0.014	0.014	0.008	0.006	0.007
Crotonaldehyde	Aldehyde	0.008	0.005	0.009	0.010	0.007	0.012	0.008	0.005	0.007
Isobutyraldehyde	Aldehyde	0.010	0.012	0.011	0.013	0.012	0.014	0.008	0.027	0.096
VOC		3.102	3.611	3.469	3.902	4.4729	4.983	3.727	2.820	3.212

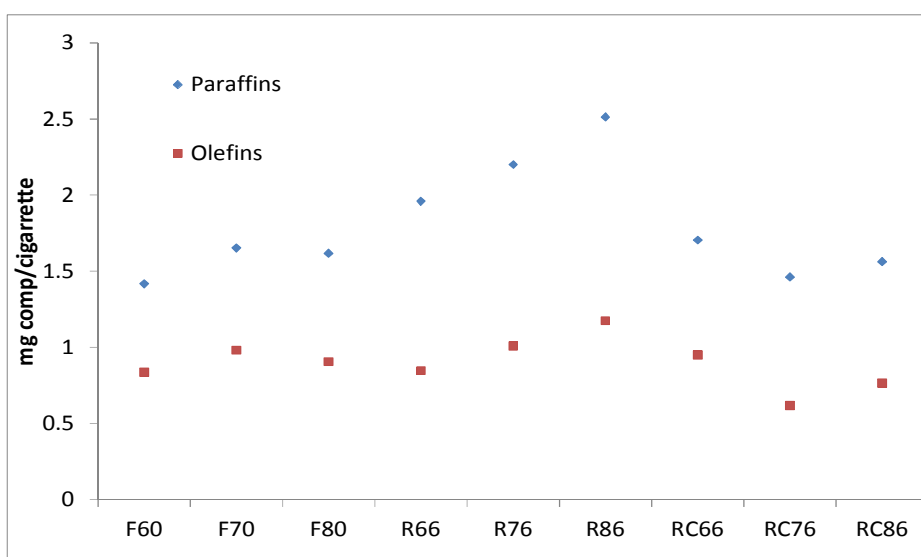


Figure 3: Yields of of paraffins and olefins appearing in the gaseous fraction of the maintream smoke from F, R and RC

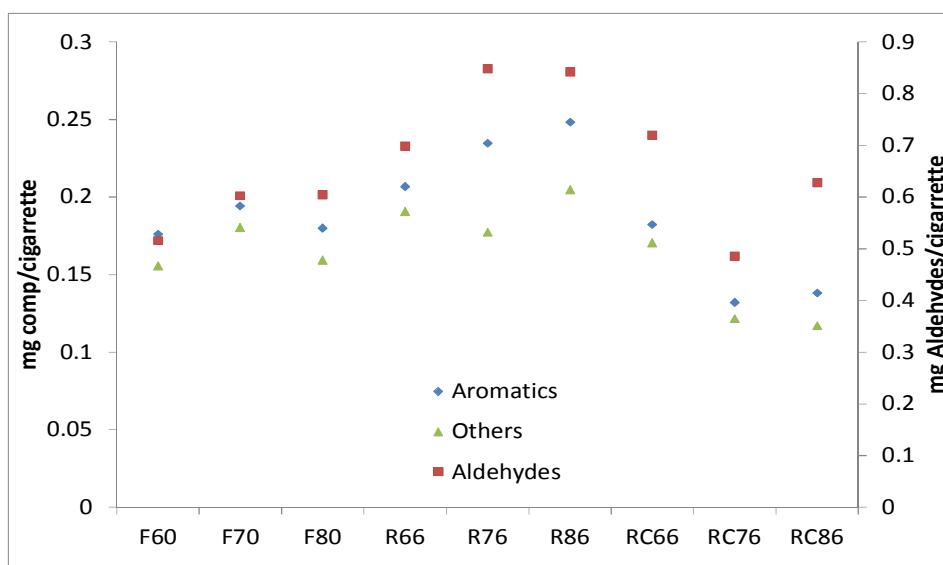


Figure 4: Yields of of aromatics, aldehydes and other compounds appearing in the gaseous fraction of the maintream smoke from F, R and RC

## Particulate Matter

The most complex fraction of the tobacco smoke is the particulate matter, also referred to in this work as liquids, condensed fraction or TPM. As was stated in the "Experimental section", there is particulate matter retained in the cigarettes' own filters (TMP-F) and in the traps located before the Tedlar bags for the gases collection (TPM-T). The results corresponding to TPM-T are especially interesting, because this fraction, together with the gases, contains the compounds that smokers inhale. Moreover, compounds appearing in TPM can condense in the mouth and the respiratory system of smoker.

Tables 4 and 5 show the yields obtained for the different compounds analyzed in TPM-F and TPM-T, respectively, from all the samples studied. As was done for the gaseous fraction, these compounds were grouped by chemical families in order to facilitate the discussion. Thus, the following groups have been considered: nitrogenous compounds, carbonylic compounds, epoxy compounds, aromatics and polyaromatic (PAH) compounds, aliphatic compounds, phenolic compounds, and others.

**Table IV:** Yield of the different compounds analyzed in the TPM-F of the mainstream smoke from F, R and RC

Compounds mg /cigarette	Family	F60	F70	F80	R66	R76	R86	RC66	RC76	RC86
Pyridine, 4-methyl-	Nitrogenous	0.003	0.004	0.003	0.006	0.010	0.007	0.005	0.002	0.002
Pyrazine, methyl-	Nitrogenous	0.002	0.004	0.003	0.003	0.005	0.004	0.004	0.002	0.002
Furfural	Carbonylic	0.030	0.043	0.034	0.043	0.055	0.037	0.034	0.021	0.020
2-Pentanone, 4-hydroxy-4-methyl-	Carbonylic	0.000	0.000	0.000	0.002	0.002	0.002	0.000	0.000	0.000
Ethanol, 2-(1-methylethoxy)-	Others	0.001	0.004	0.003	0.002	0.003	0.003	0.003	0.002	0.002
2-Furanmethanol	Epoxy	0.005	0.011	0.010	0.009	0.015	0.009	0.009	0.005	0.004
Pyridine, 3-methyl-	Nitrogenous	0.003	0.008	0.007	0.009	0.017	0.014	0.008	0.004	0.004
2-Propanone, 1-(acetyloxy)-	Carbonylic	0.010	0.015	0.012	0.015	0.020	0.013	0.010	0.006	0.006
4-Cyclopentene-1,3-dione	Carbonylic	0.009	0.014	0.012	0.011	0.015	0.010	0.011	0.006	0.006
Styrene	Aromatic	0.001	0.001	0.001	0.001	0.003	0.003	0.002	0.001	0.001
2-Cyclopenten-1-one, 2-methyl-	Carbonylic	0.011	0.017	0.014	0.016	0.023	0.018	0.013	0.008	0.008
2-Acetylfuran	Carbonylic	0.004	0.008	0.007	0.012	0.015	0.009	0.008	0.005	0.004
2(5H)-furanone	Carbonylic	0.008	0.011	0.009	0.010	0.016	0.007	0.006	0.004	0.004
Pyrazine, 2,3-dimethyl-	Nitrogenous	0.001	0.002	0.001	0.002	0.002	0.001	0.001	0.001	0.001
2-Hydroxycyclopent-2-en-1-one	Carbonylic	0.003	0.006	0.005	0.008	0.010	0.007	0.005	0.003	0.003
Pyridine, 3,5-dimethyl-	Nitrogenous	0.000	0.003	0.002	0.004	0.003	0.002	0.004	0.002	0.002
2,5-Dimethyl-2-cyclopentenone	Carbonylic	0.002	0.003	0.002	0.002	0.003	0.003	0.003	0.002	0.002
2(3H)-furanone, 5-methyl-	Carbonylic	0.001	0.002	0.001	0.002	0.003	0.002	0.001	0.001	0.001
Butanoic acid, 3-methyl-	Others	0.006	0.003	0.003	0.001	0.003	0.001	0.003	0.001	0.001
Ethanol, 2-butoxy-	Others	0.001	0.002	0.001	0.003	0.003	0.001	0.002	0.001	0.001
Benzaldehyde	Carbonylic	0.003	0.005	0.004	0.006	0.009	0.008	0.006	0.003	0.003
Furfural, 5-methyl-	Carbonylic	0.017	0.026	0.022	0.023	0.031	0.020	0.018	0.010	0.008
Pyridine, 3-ethenyl-	Nitrogenous	0.001	0.006	0.005	0.001	0.004	0.003	0.003	0.002	0.002
2(5H)-Furanone, 3-methyl-	Carbonylic	0.003	0.004	0.003	0.005	0.006	0.005	0.005	0.002	0.003
Phenol	Phenolic	0.024	0.040	0.034	0.039	0.053	0.040	0.032	0.019	0.019
2-isopropylfuran	Epoxy	0.002	0.007	0.005	0.006	0.010	0.008	0.006	0.003	0.003
2-Cyclopenten-1-one, 2-hydroxy-3-methyl-	Carbonylic	0.007	0.017	0.013	0.019	0.027	0.020	0.013	0.010	0.008
Limonene	Others	0.005	0.007	0.008	0.012	0.020	0.018	0.015	0.011	0.009
Benzenemethanol	Aromatics	0.007	0.011	0.011	0.000	0.004	0.000	0.003	0.002	0.002
2,3-Dimethyl-2-cyclopenten-1-one	Carbonylic	0.004	0.007	0.006	0.009	0.014	0.011	0.006	0.004	0.003
Indeno	PAH	0.001	0.005	0.002	0.005	0.006	0.006	0.004	0.002	0.002
o-Cresol	Phenolic	0.010	0.019	0.016	0.024	0.031	0.020	0.027	0.009	0.008
2-Acetylpyrrole	Nitrogenous	0.002	0.003	0.003	0.004	0.005	0.005	0.003	0.002	0.002
Phenol, 4-methoxy-	Phenolic	0.001	0.002	0.001	0.002	0.003	0.003	0.002	0.001	0.001
Ethanone, 1-phenyl-	Carbonylic	0.001	0.002	0.002	0.003	0.002	0.003	0.001	0.001	0.001
p-Cresol	Phenolic	0.017	0.029	0.025	0.029	0.053	0.029	0.021	0.014	0.013
2 ethyl tiophene	Others	0.002	0.005	0.003	0.005	0.006	0.003	0.003	0.002	0.002

Table IV: Yield of the different compounds analyzed in ... (Cont)

Phenol, 2-methoxy-	Phenolic	0.007	0.012	0.011	0.014	0.018	0.014	0.011	0.006	0.006
2-Propanamine	Nitrogenous	0.002	0.005	0.004	0.006	0.009	0.005	0.005	0.002	0.002
3-Ethyl-2-hydroxy-2-cyclopenten-1-one	Carbonylic	0.004	0.007	0.006	0.011	0.015	0.011	0.006	0.003	0.002
Benzeneacetonitrile	Nitrogenous	0.003	0.006	0.005	0.005	0.007	0.006	0.004	0.002	0.002
2,3-Dihydro-3,5-dihydroxy-6-methyl-4H-pyran-4-one	Carbonylic	0.005	0.009	0.008	0.003	0.003	0.003	0.003	0.002	0.002
Phenol, 2,4-dimethyl-	Phenolic	0.006	0.008	0.006	0.003	0.011	0.003	0.000	0.000	0.000
Phenol, 4-ethyl-	Phenolic	0.004	0.006	0.006	0.005	0.008	0.002	0.000	0.000	0.000
Naphthalene	PAH	0.001	0.002	0.002	0.004	0.007	0.003	0.004	0.002	0.002
Ethanone, 1-(3-methylphenyl)-	Carbonylic	0.002	0.003	0.003	0.002	0.005	0.002	0.001	0.001	0.001
p-cresol 2 methoxy	Phenolic	0.001	0.001	0.001	0.002	0.003	0.002	0.002	0.001	0.001
2,3-Dihydro-benzofuran	Epoxy	0.003	0.005	0.004	0.005	0.016	0.006	0.006	0.003	0.003
2-furancarboxaldehyde, 5-(hydroxymethyl)-	Carbonylic	0.002	0.003	0.003	0.010	0.013	0.008	0.003	0.003	0.000
1H-Inden-1-one, 2,3-dihydro-	Carbonylic	0.003	0.005	0.005	0.007	0.009	0.008	0.005	0.002	0.002
Hydroquinone	Phenolic	0.001	0.012	0.011	0.020	0.009	0.025	0.021	0.015	0.017
1H-Indole	Nitrogenous	0.009	0.019	0.016	0.009	0.028	0.005	0.006	0.015	0.004
4-vinyl-2-methoxy-phenol	Phenolic	0.004	0.004	0.004	0.007	0.008	0.009	0.008	0.003	0.003
Nicotine	Nitrogenous	0.423	0.618	0.559	0.793	1.007	0.793	0.635	0.487	0.470
1H-Indole, 3-methyl-	Nitrogenous	0.004	0.004	0.005	0.007	0.011	0.009	0.007	0.004	0.004
Myosmine	Nitrogenous	0.003	0.008	0.007	0.006	0.010	0.008	0.006	0.005	0.005
Phenol, 2-methoxy-4-(2-propenyl)-	Phenolic	0.001	0.004	0.002	0.007	0.013	0.009	0.007	0.004	0.005
Nicotyrine	Nitrogenous	0.004	0.005	0.006	0.010	0.011	0.009	0.006	0.005	0.005
Norsolanadiona	Carbonylic	0.002	0.002	0.001	0.002	0.003	0.002	0.004	0.003	0.003
2,3'-Bipyridine	Nitrogenous	0.003	0.007	0.006	0.009	0.013	0.009	0.009	0.007	0.007
1,4-dihydrophenanthrene	PAH	0.001	0.001	0.002	0.003	0.005	0.004	0.000	0.000	0.000
Megastigmatrienone	Carbonylic	0.001	0.002	0.003	0.003	0.005	0.004	0.004	0.004	0.003
N-propyl- nornicotine	Nitrogenous	0.000	0.000	0.000	0.002	0.004	0.002	0.000	0.000	0.000
Cotinine	Nitrogenous	0.000	0.007	0.007	0.005	0.009	0.009	0.008	0.004	0.004
1H-Indene, 2,3-dihydro-1,1,3-trimethyl-3-phenyl-	Aromatic	0.000	0.000	0.001	0.001	0.002	0.002	0.002	0.002	0.002
5-Tetradecene	Aliphatic	0.001	0.002	0.002	0.000	0.000	0.000	0.000	0.000	0.000
N(b)-formylnornicotine	Nitrogenous	0.001	0.003	0.002	0.002	0.004	0.004	0.002	0.000	0.000
2,4-Diphenyl-4-methyl-penten-1ene	Aromatic	0.001	0.001	0.001	0.001	0.001	0.000	0.001	0.001	0.001
Neophytadiene	Aliphatic	0.021	0.042	0.038	0.069	0.088	0.077	0.066	0.044	0.047
Farnesol	Others	0.001	0.003	0.003	0.003	0.005	0.005	0.001	0.001	0.001
8-Quinolinemethanol	Nitrogenous	0.000	0.001	0.001	0.004	0.003	0.005	0.009	0.003	0.009
Hexadecanoic acid, ethyl ester	Others	0.001	0.003	0.003	0.002	0.007	0.007	0.006	0.006	0.006
Eicosane	Aliphatic	0.002	0.003	0.003	0.001	0.002	0.001	0.001	0.001	0.001
pentadecane	Aliphatic	0.001	0.002	0.002	0.001	0.001	0.002	0.001	0.001	0.001
Docosane	Aliphatic	0.000	0.000	0.000	0.001	0.001	0.002	0.001	0.001	0.000
Tricosane	Aliphatic	0.002	0.007	0.007	0.009	0.013	0.012	0.005	0.003	0.004
2,6,10,14,18,22-Tetracosahexaene, 2,6,10,15,19,23-hexamethyl-	Aliphatic	0.001	0.004	0.003	0.002	0.003	0.002	0.005	0.002	0.001
Heptacosane	Aliphatic	0.002	0.006	0.006	0.007	0.011	0.010	0.005	0.003	0.004
Triacotane	Aliphatic	0.002	0.005	0.006	0.007	0.012	0.011	0.002	0.001	0.002
Octadecane	Aliphatic	0.004	0.014	0.013	0.020	0.029	0.028	0.012	0.006	0.008
Tocopherol	Phenolic	0.001	0.004	0.005	0.007	0.015	0.015	0.005	0.002	0.003
Total		0.759	1.232	1.093	1.454	1.971	1.512	1.201	0.835	0.816





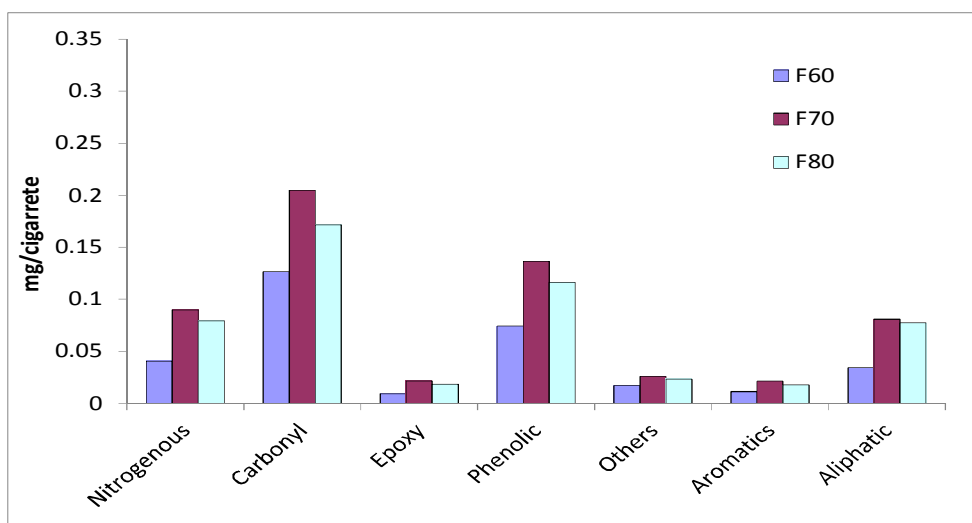
Table V: Yield of the different compounds analyzed in ... (Cont)

p-cresol 2 methoxy	Phenolic	0.005	0.003	0.002	0.004	0.004	0.002	0.000	0.000	0.000
2,3-Dihydro-benzofuran	Epoxy	0.004	0.003	0.003	0.001	0.002	0.002	0.001	0.000	0.000
2-furancarboxaldehyde, 5-(hydroxymethyl)-	Carbonylic	0.005	0.002	0.001	0.005	0.004	0.003	0.001	0.000	0.000
1H-Inden-1-one, 2,3-dihydro-	Carbonylic	0.003	0.000	0.000	0.002	0.002	0.001	0.000	0.000	0.000
Hydroquinone	Phenolic	0.018	0.018	0.017	0.012	0.012	0.011	0.004	0.002	0.002
1H-Indole	Nitrogenous	0.009	0.006	0.005	0.005	0.006	0.003	0.008	0.007	0.007
4-vinyl-2-methoxy-phenol	Phenolic	0.008	0.004	0.004	0.005	0.005	0.004	0.001	0.001	0.001
Nicotine	Nitrogenous	1.046	0.750	0.677	0.609	0.650	0.558	0.357	0.229	0.216
1H-Indole, 3-methyl-	Nitrogenous	0.006	0.004	0.003	0.002	0.003	0.002	0.001	0.001	0.001
Myosmine	Nitrogenous	0.009	0.005	0.005	0.004	0.004	0.004	0.002	0.001	0.001
Phenol, 2-methoxy-4-(2-propenyl)-	Phenolic	0.007	0.003	0.011	0.002	0.002	0.002	0.002	0.001	0.001
Nicotyrine	Nitrogenous	0.010	0.006	0.006	0.004	0.004	0.004	0.002	0.001	0.001
Norsolanadiona	Carbonylic	0.001	0.003	0.003	0.001	0.001	0.001	0.001	0.001	0.001
2,3'-Bipyridine	Nitrogenous	0.009	0.006	0.006	0.004	0.006	0.006	0.002	0.001	0.001
1,4-dihydrophenanthrene	PAH	0.005	0.002	0.002	0.001	0.004	0.001	0.000	0.000	0.000
Megastigmatrienone	Carbonylic	0.004	0.003	0.003	0.002	0.002	0.002	0.001	0.001	0.001
N-propyl- nornicotine	Nitrogenous	0.000	0.000	0.000	0.001	0.002	0.001	0.000	0.000	0.000
Cotinine	Nitrogenous	0.012	0.007	0.008	0.007	0.006	0.007	0.003	0.002	0.002
1H-Indene, 2,3-dihydro-1,1,3-trimethyl-3-phenyl-	Aromatic	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
5-Tetradecene	Aliphatic	0.003	0.003	0.002	0.002	0.000	0.002	0.001	0.001	0.001
N(b)-formylnornicotine	Nitrogenous	0.005	0.003	0.003	0.004	0.005	0.004	0.002	0.001	0.002
2,4-Diphenyl-4-methyl-penten-1ene	Aromatic	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Neophytadiene	Aliphatic	0.057	0.040	0.037	0.039	0.042	0.037	0.026	0.014	0.014
Farnesol	Others	0.004	0.004	0.003	0.002	0.003	0.003	0.001	0.000	0.000
8-Quinolinemethanol	Nitrogenous	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.003	0.003
Hexadecanoic acid, ethyl ester	Others	0.002	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Eicosane	Aliphatic	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000
pentadecane	Aliphatic	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Docosane	Aliphatic	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Tricosane	Aliphatic	0.008	0.006	0.005	0.005	0.007	0.007	0.005	0.002	0.002
2,6,10,14,18,22-Tetracosahexaene, 2,6,10,15,19,23-hexamethyl-	Aliphatic	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Heptacosane	Aliphatic	0.007	0.006	0.005	0.005	0.006	0.006	0.003	0.002	0.002
Triacotane	Aliphatic	0.007	0.006	0.006	0.005	0.006	0.007	0.002	0.001	0.001
Octadecane	Aliphatic	0.020	0.017	0.014	0.013	0.017	0.017	0.006	0.003	0.004
Tocopherol	Phenolic	0.008	0.005	0.006	0.005	0.008	0.010	0.003	0.000	0.001
Total		1.387	0.986	0.913	0.809	0.878	0.755	0.466	0.293	0.283

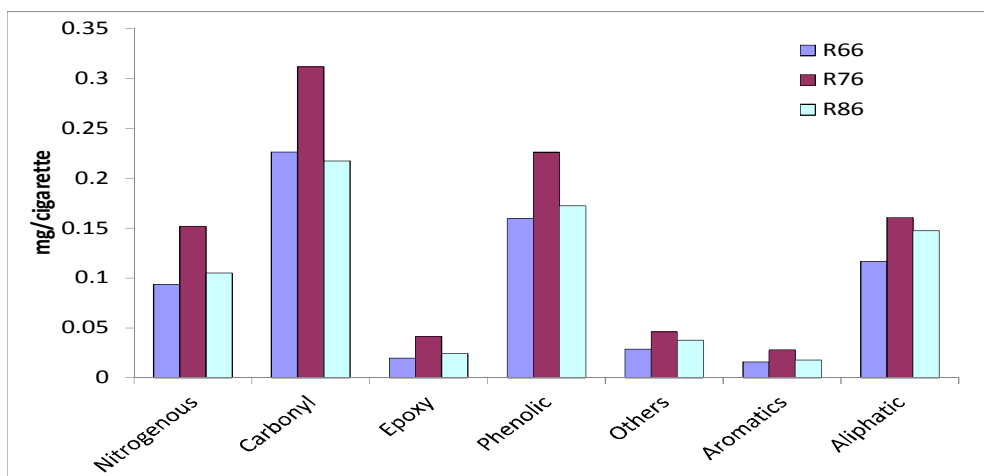
According to Table IV, the sums of yields obtained for the different compounds analyzed in the TPM-F fraction from F and RC follow the same trend as the global amounts shown in Table II, that have been already discussed. Nevertheless, the case of R is quite different, and whereas the amount of TMP-F decreases as WTC increases, the sum of the yields of the different compounds analyzed passes through a maximum. This difference enhances the existence of compounds that appear in the mainstream smoke, but that have not been analyzed in the present work. This fact is obvious, as the mere comparison between the TPM-F values of Table II and the total sums of Table IV points out and enhances the need of carrying out the analysis of results through the comparison compound by compound or, much better, by grouping compounds by chemical families. Nicotine is, by far, the major compound appearing in all the cases. Nevertheless, other compounds are formed in important quantities such as, for

example, neophytadiene, furfural, p-cresol and phenol. As has already been commented, the catalyst produces a significant decrease of practically all compounds at all WTC. In fact, many compounds are practically not detected in the case of RC, as for example 4-hydroxy-4-methyl-2-pentanone, some phenol derivatives, 1,4-dihydrophenanthrene or N-popyl-nornicotine among others. The reduction observed for nicotine, that represents more than 50% of the total mass of compounds analyzed in filters, is in the range of 20-51%, being the highest value for the sample with the intermediate value of WTC.

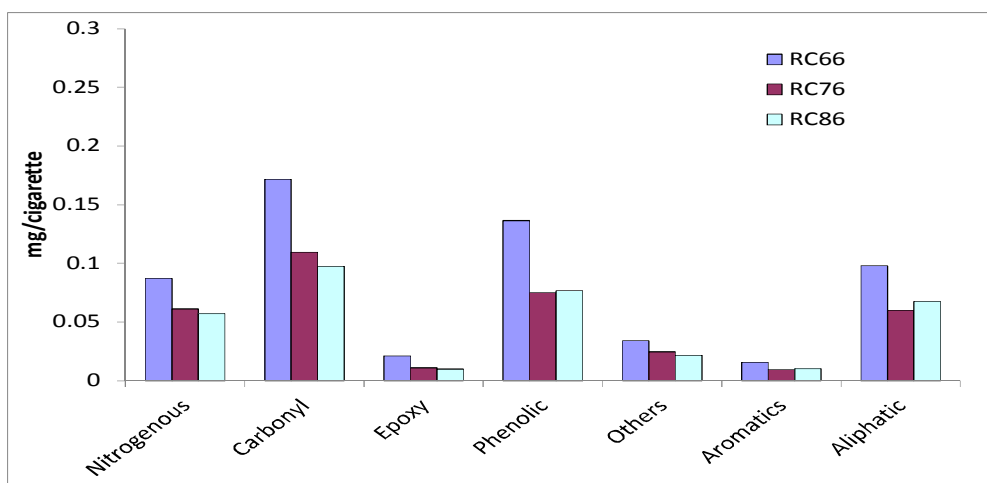
Figure 5 shows the yields obtained for the different chemical families of compounds appearing in TPM-F, at each WTC value, for F (Figure 5a), R (Figure 5b) and RC (Figure 5c) cigarettes. In this figure, nicotine has been excluded from the group of nitrogenous compounds in order to avoid the distortion associated with the high yields of nicotine obtained in all the cases, that has already been commented on. It can be observed that the highest yields from both tobaccos, F and R, correspond to the intermediate WTC (Figures 5a and 5b). Moreover, if nicotine is not taken into account, the chemical family providing the largest yields corresponds, in both cases, to the carbonyl compounds. The decreasing order of yields of the different groups obtained from F is as follows: carbonyl, phenolic, nitrogenous, aliphatic, others, aromatic and epoxy compounds, whereas the decreasing order of groups from R and RC is: carbonyl, phenolic, aliphatic, nitrogenous, others, epoxy and aromatic compounds. Moreover, the yield of all the families is higher for R than for F tobacco. The noticeable effect of the catalyst can be seen in Figure 5c, where the trends observed are different than those previously commented on for R and F cigarettes. In fact, in the presence of catalyst, the obtained yields of the chemical families studied decrease as WTC increases. Moreover, interesting reductions have been obtained, especially for the carbonyls group, and being nitrogenous compounds (nicotine excluded) the least affected. The effect of the catalyst is especially significant at the two higher WTC tested, where reductions higher than 50% can be observed for all the groups.



**Figure 5a:** Yield of the different chemical families of compounds appearing in TPM-F obtained when smoking F cigarettes. Nicotine has not been counted in the nitrogenous compounds group.



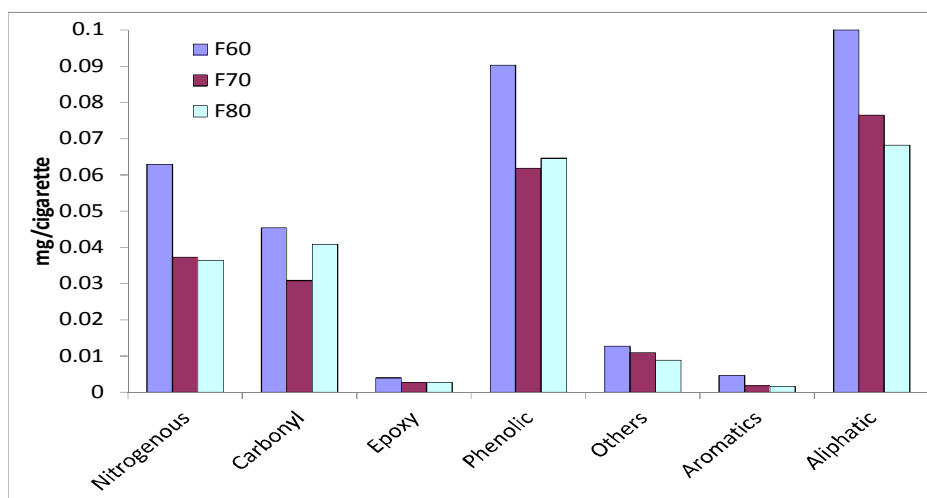
**Figure 5b:** Yield of the different chemical families of compounds appearing in TPM-F obtained when smoking R cigarettes. Nicotine has not been counted in the nitrogenous compounds group.



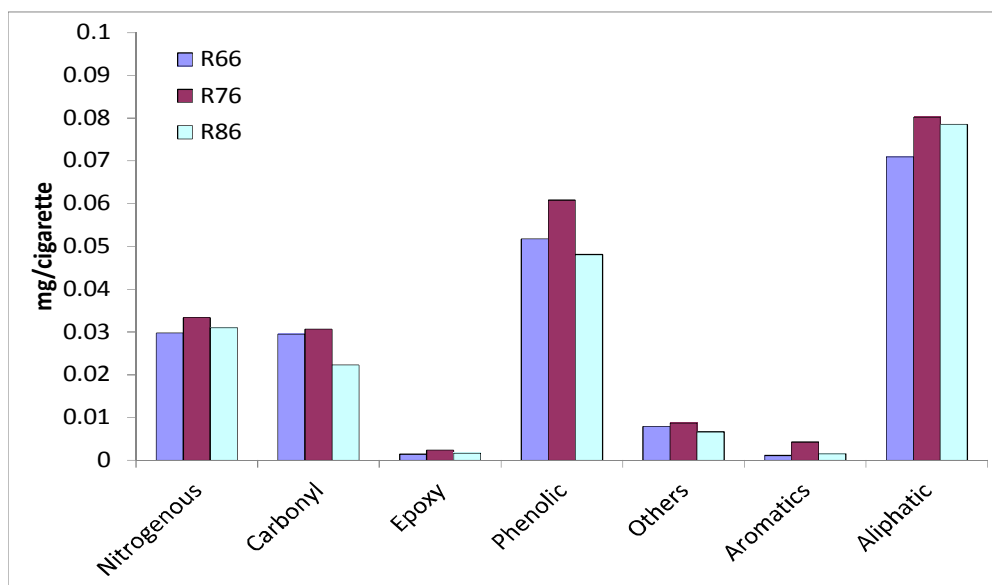
**Figure 5c:** Yield of the different chemical families of compounds appearing in TPM-F obtained when smoking RC cigarettes. Nicotine has not been counted in the nitrogenous compounds group.

Table V shows the results corresponding to the compounds analyzed in the fraction of the mainstream smoke that was retained in traps (TPM-T). As has already been commented, this fraction is especially interesting because it contains compounds that are inhaled by smokers and can condense in the mouth and lung. As can be seen, Contrary to the behaviour observed in the filters, in traps more compounds are retained in the case of F tobacco, and this effect may be due to the fact that filters of F cigarettes are smaller than R filters. In general, the total mass of compounds analyzed in TPM-T is lower than that of TPM-F, the case of F60 being the only exception. As in the case of TPM-F, the major compound retained in TPM-T is nicotine, and some materials, such as hydroquinone, also appear in significant quantities. However, several compounds that are retained in the filters are not detected in the traps, as is the case of furfural and benzaldehyde, among others. As in the previous cases, when the catalyst was added important reductions in most of the compounds can be observed, and the majority of the compounds are not detected. This reduction increases, in general, when the WTC increases. Finally, with respect to the nicotine, that in the traps represents more than 75% of the total of condensed products, the largest reduction obtained was around 65%, at the intermediate WTC.

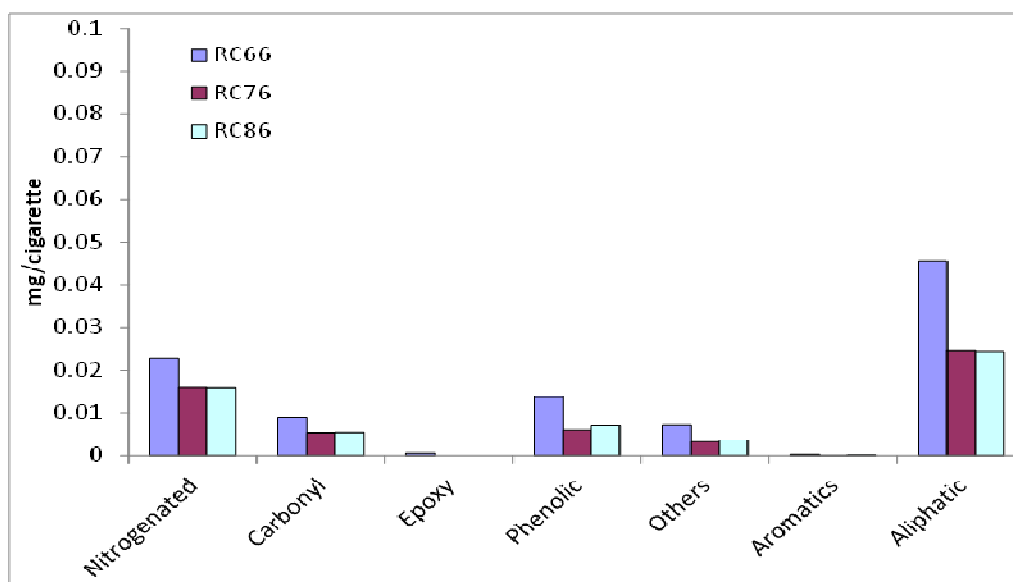
Figure 6 shows the yield of the same chemical families considered in the previous paragraphs in the TPM-T from F, R and RC cigarettes. As it can be seen, F and RC cigarettes behave in a similar way, showing a clear decreasing trend when WTC increases (Figures 6a and 6c). However, R presents a similar tendency as in filters, and a maximum can be observed at the intermediate WTC value (Figure 6b). In the case of F and R tobaccos, the major family retained in traps corresponds to aliphatic compounds, followed by phenolic compounds. Again, when the catalyst has been added to tobacco, the reductions obtained in all the families are very important, especially for aromatic compounds that practically disappear. In the RC case, the most abundant family is, as for tobaccos, the aliphatic group, but, the second most abundant family is the nitrogenous compounds group (with nicotine excluded).



**Figure 6a:** Yield of the different chemical families of compounds appearing in TPM-T obtained when smoking F cigarettes. Nicotine has not been counted in the nitrogenous compounds group.



**Figure 6b:** Yield of the different chemical families of compounds appearing in TPM-T obtained when smoking R cigarettes. Nicotine has not been counted in the nitrogenous compounds group.



**Figure 6c:** Yield of the different chemical families of compounds appearing in TPM-T obtained when smoking RC cigarettes. Nicotine has not been counted in the nitrogenous compounds group.

Table VI shows the reductions obtained in the yields of the different chemical families analyzed in TPM-F and TPM-T as a consequence of the use of the catalyst as additive for the reference cigarette. The percentage of reduction with respect to the reference tobacco has been calculated as follows:

$$\% \text{ reduction} = \frac{\text{yield in families in R tobacco} - \text{yield in families in RC tobacco}}{\text{yield in families in R tobacco}} \cdot 100$$

For this comparison, nicotine has been included in the nitrogenous compounds group. It can be observed that in both cases, filters and traps, the catalyst produces very noticeable reductions in all the families of compounds, with the only exception of the groups of others, epoxies and aromatics being at the lowest WTG value. This reductive effect is more important at the medium WTC value, which is the one used in the commercial cigarettes. Moreover, the reductions obtained in traps are higher than those obtained in filters. The results shown in Table VI have been normalized with respect to nicotine that is the most abundant compound, as many authors do in order to compare and discuss their results<sup>[24]</sup>, and the results obtained are shown in Table VII.

For the normalization, the yield obtained for the different families has been divided for the yield of the nicotine, and then, the reduction was calculated as in Table VI. Though these data may be subjected to high dispersion levels, it could be concluded that the catalyst is capable of a selective reduction effect with respect to the nicotine (that is also highly reduced), that depends on the WTC, this effect being more noticeable at the intermediate WTC values, that are those used in commercial cigarettes. The chemical family that presents higher reductions than nicotine in all the cases studied is the group of carbonyls, especially in TPM-T. If neither the groups of nitrogenous compounds that, obviously, have lost significance once nicotine has been discounted, nor other compounds are taken into account, in TPM-T, only aliphatics at the lowest WTC value show lower reductions than nicotine. However, in TPM-F, at the lowest WTC value, aromatic, phenolics and epoxies, besides aliphatics are also less reduced than nicotine.

**Table VI:** Reduction of the yields of different families of chemical compounds analyzed in the TPM retained in filters and traps. Nicotine is included in the nitrogenous compounds group. Reductions represent the difference between the yields obtained from R and RC cigarettes, expressed as percentage of the yields obtained from R.

Filters	Nitrogenous	Carbonyl	Epoxy	Phenolic	Other	Aromatic	Aliphatic
RC66	18.49	24.14	-7.04	15.33	-18.31	-0.20	16.06
RC76	60.66	52.85	73.36	67.89	46.64	65.71	62.69
RC86	41.51	55.11	58.90	57.40	41.51	41.46	54.11
<b>Traps</b>							
RC66	40.57	69.73	54.25	70.75	7.82	68.91	35.60
RC76	64.14	82.02	100.00	91.05	62.66	98.97	69.13
RC86	60.64	75.04	100.00	85.22	43.59	87.60	68.77

**Table VII:** Reduction of the yields of different families of chemical compounds analyzed in the TPM, normalized with respect to nicotine.

Filters	Nitrogenous	Carbonyl	Epoxy	Phenolic	Other	Aromatic	Aliphatic
RC66	-1.69	5.37	-33.54	-5.62	-47.59	-24.99	-4.71
RC76	2.49	27.32	44.92	33.59	-10.34	29.09	22.84
RC86	1.36	24.29	30.68	28.16	1.35	1.28	22.62
<b>Traps</b>							
RC66	-1.36	48.37	21.98	50.12	-57.21	46.98	-9.84
RC76	-1.67	49.04	100.00	74.63	-5.87	97.07	12.47
RC86	-1.79	35.43	100.00	61.77	-45.89	67.93	19.22

The comparison of the results obtained in this work with the data reported by other authors is difficult because only two studies considering the influence of the weight of tobacco per cigarette (WTC) on the smoke chemistry have been found [8, 10], and in both cases, other cigarette design parameters have also been modified. In the work of Darrall and Figgins<sup>[10]</sup>, CO, tar and nicotine were measured as a function of WTC, but the cigarette diameter was not kept constant. Despite the results showing a wide dispersion, some linear increasing trend with WTC was observed. Additionally these authors [10] reported a significant influence of the type of tobacco blend. Nevertheless, they studied a much wider WTC interval and a closer inspection of their results in the zone we have covered in our study show a decreasing trend, since they describe a maximum. Our results belong to the larger end of the interval covered by these authors and show different trends depending on the tobacco blend smoked, the mainstream smoke fraction considered and the presence of a catalyst. Dagnon et al.<sup>[8]</sup> studied the presence of phenols in mainstream tobacco smoke and analyzed the effect of the amount of tobacco per cigarette, concluding that the amount of phenols decreased when the amount of tobacco smoked also decreased. They also found a strong dependence on the variety of tobacco.

No papers have been found in the literature on the effect of WTC when using a catalyst; consequently no reference can be used to compare other trends.

## CONCLUSIONS

The results obtained in this work reflect a significant influence of the weight of tobacco per cigarette (WTC) on the smoke chemistry, despite any uniform tendency, which could be considered as a general rule, can be established. The first factor that

causes differences in the observed trends is the type of tobacco blend smoked, as the differences in the results obtained for R and F cigarettes reflect. In general, the following trends have been found:

- The amount of tobacco smoked increases as WTC increases in all the cases.
- The amount of TPM-T and TPM-F decreases as WTC increases for R and RC, and passes through a maximum at the intermediate value of WTC for F.
- The CO yield shows a maximum at the intermediate value of WTC for F, and a minimum for RC, whereas it increases as WTC also increases for R.
- As WTC increases, the total VOCs obtained from F pass through a maximum, increase in the case of R, and pass through a minimum in the case of RC.
- The different families of chemical compounds analyzed in the TPM-F fraction of the mainstream smoke show a maximum at the intermediate value of WTC for R and F cigarettes but seem to decrease as WTC increases when the catalyst has been added to C cigarette.
- The different families of chemical compounds analyzed in the TPM-T fraction of the mainstream smoke show a minimum at the intermediate value of WTC for F and RC cigarettes and a maximum for R cigarettes.

The major compounds of the gaseous fraction of the mainstream smoke are paraffins and the major compound of the particulate matter is nicotine, followed by carbonylic compounds in the case of TPM-F, and by aliphatic and phenolics in the case of TPM-F. The presence of the catalyst causes noticeable selective reductions in the main part of compounds and mainstream fractions considered.

## Acknowledgements

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