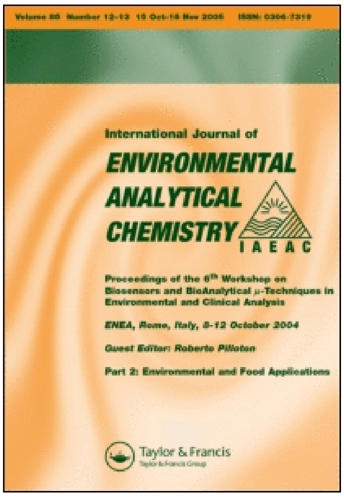

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Multi-Element Analysis of Tobacco and Smoke Condensate by Instrumental Neutron Activation Analysis and Atomic Absorption Spectrometry

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MULTI-ELEMENT ANALYSIS OF TOBACCO AND SMOKE CONDENSATE BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS AND ATOMIC ABSORPTION SPECTROMETRY

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The tobacco and tobacco smoke of the brands *Marlboro* and the average cigarette *C-20* have been analysed for 23 elements. The tobacco smoke condensate TSC was collected under standard conditions in a quartz tube by electrostatic precipitation using a commercial smoking machine. Instrumental neutron activation analysis was used for the detection of the elements Ag, As, Au, Br, Cd, Ce, Co, Cr, Fe, Hg, K, Mn, Na, Ni, Sb, Se, Th, U and Zn in tobacco and TSC, and atomic absorption spectrometry for the elements As, Cd, Cr, Cu, Fe, K, Mn, Na, Ni, Pb, Tl and Zn. For analysis by atomic absorption spectrometry, the tobacco and TSC samples were digested in a pressure microwave decomposition system. The results of the two methods showed good agreement. The contents determined in the smoke condensate were lower than 1 ng/cig. for the elements Ag, Au, Ce, Co, Cr, Fe, Hg, Sb, Sc, Th, Tl and U; for the elements As, Cd, Cu, Mn, Ni, Pb, Se and Zn, they were between 1 and 100 ng/cig., and for the elements Br, K and Na, they were above 100 ng/cig. For the most relevant elements, the percentage transfer from the cigarette tobacco into the smoke was estimated. The results are compared with those presented in the literature.

KEY WORDS: Tobacco, tobacco smoke, trace element analysis, electrostatic precipitation.

INTRODUCTION

Several inorganic and metallic constituents of cigarette smoke are suspected to be carcenogenic or toxic even at very low levels of intake and may have synergistic interactions with other harmful compounds such as nicotin, benzpyrene, CO, NO_x, H_2S, nitrosamines and many others of the about 4,000 substances determined in the smoke condensate[1]. Therefore, the analysis of tobacco smoke for trace elements is of great importance. The content of toxic elements in cigarette tobacco is the result of the uptake of these elements by the tobacco plants from the soil and the atmosphere. Because tobacco leaves are not washed before drying[2], the deposition of atmospheric dust on the surface of the tobacco leaves can, in the case of heavy metals such as cadmium and lead, essentially contribute to their content in the cigarette[2,3,4].

The toxic elements incorporated by inhaling the cigarette smoke are distributed in the organism and/or enriched in some organs. Cadmium contents were found to be two to four times higher in lungs and kidneys of smokers compared with those of non-smokers[5,6,7].

One has to differentiate between two kinds of smoke with a different content of toxic substances, i.e., the mainstream smoke and the sidestream smoke (see Figure 1). The mainstream smoke is directly inhaled by the smoker. The temperatures in the burning zone of the cigarette are about 700–900°C[8]. During smoking, many of the leaf components are destroyed, some are distilled over, and a large number of new compounds is formed. Non-volatile compounds are mainly retained in the filter, whereas the volatile substances pass into the mainstream. Because of lower temperatures during its formation, the composition of the sidestream smoke is different from that of the mainstream.

Several investigations have been reported on the determination of toxic trace elements in tobacco and in tobacco smoke[2,10–13, 15–50]. However, in most cases, only small groups of elements have been determined. In general, the results obtained for the individual elements in different investigations show large deviations. Therefore, there is a need for more comprehensive investigations of the elemental composition of cigarette smoke.

In the present work, tobacco and smoke condensate of two cigarette brands (Marlboro and the average cigarette C-20) were analysed by using instrumental neutron activation analysis (INAA) and atomic absorption spectrometry (AAS) for 23 elements.

EXPERIMENTAL SECTION

Cigarettes

The cigarette brands investigated were *Marlboro* (Philip Morris, Munich, Germany) and the average cigarette of the German market *C-20* (Verband der Tabakindustrie, Bonn, Germany).

Reagents

All reagents used were of *pro analysi* quality. Nitric acid (p. a., 65%; Merck, Darmstadt, Germany) was purified by subboiling distillation using a quartz apparatus. The water used was obtained by double distillation. Calibration solutions with analyte concentrations of 1 g/l were prepared by diluting stock standard solutions by Merck.

Apparatus

A commercially available smoking machine RM 20/CS (H. Borgwaldt, Hamburg, Germany) was used for smoking the cigarettes under standard conditions[9,10].

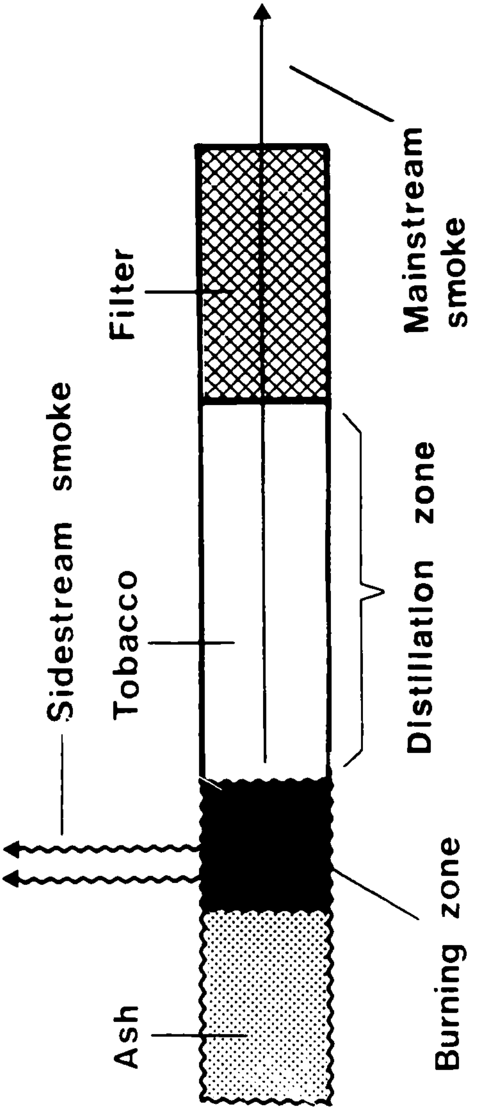

Figure 1 Schematic illustration of the distribution of the elements during cigarette smoking.

Dry tobacco was homogenized in a Microdismembrator II (Braun, Melsungen, Germany).

A pressure microwave decomposition (PMD) system (Kürner, Rosenheim, Germany) with quartz bombs was used for mineralization of tobacco and tobacco smoke condensate.

A Perkin-Elmer atomic absorption spectrometer Model Zeeman-5000 with a graphite furnace HGA-500 and an autosampling system AS-70 was used for the determination of Cd, Cu, Mn, Ni, Pb and Zn, and a Perkin-Elmer spectrometer Zeeman-4100 ZL with transversal heated graphite furnace (THGA) and longitudinal Zeeman effect for the determination of As, Cr, Mn and Tl. Both spectrometer systems were equipped with electrodeless discharge lamps (EDL) for As, Pb, Tl and hollow cathode lamps (HCL) for the other elements. A Perkin-Elmer Model 400 atomic absorption spectrometer was used to determine Na and K with the flame technique.

A high-resolution γ-spectrometer system (EG&G Ortec, Munich, Germany) consisting of an intrinsic germanium detector with an efficiency of 44% relative to a 3 × 3 in. Na(Tl)-detector, a fwhm of 1.72 keV at the 1332-keV γ-ray of ^{60}Co and a peak-to-Compton ratio of 78:1 and an ADCAM multichannel buffer was used for counting the indicator radionuclides. The evaluation of the γ-ray spectra was carried out with the DSM computer 80286 by using the software package "Omnigam" (EG&G Ortec).

Smoke condensate collection and sample preparation

The particulate matter of the smoke was collected on the walls of a quartz tube in an electric field of 25–18 kV using an electrode made of high purity aluminium. Filter cigarettes of the brands *C-20* and *Marlboro* were taken from commercially available packets, but cigarettes with a deviation from the average weight of more than ± 20 mg were rejected[10]. Prior to smoking, the cigarettes were conditioned for three days at room temperature in a closed box made of polyethylene at a humidity of 60% achieved by using a saturated solution of ammonium nitrate. The conditioned 85-mm filter cigarettes were automatically smoked to a butt length of 23 mm under standard conditions (35 ml puffs of 2 sec duration each minute)[9,10] using the smoking machine RM 20/CS with its outlet pipe connected directly to the electrostatic trap. The original glass tubes were replaced by quartz tubes. Before use, the quartz tubes were cleaned by washing with purified nitric acid (65%). After smoking of 100 cigarettes, the smoke condensate was taken from the quartz tube with polysterene stirring rods and kept in polysterene vessels for analysis by AAS and INAA.

AAS

About 100 mg of the smoke condensate were weighed in a quartz tube of the pressure microwave decomposition system. The samples were digested in 2 ml HNO_3 at step 8 for 10 min.

The tobacco samples were dried in an oven for 5 h at 100°C, homogenized using the microdismembrator and kept for 12 h in a desiccator. About 100-mg portions of the dry tobacco powders were digested under the same conditions as given for smoke condensate.

Table 1 Instrumental and experimental conditions used for GFAAS.

Parameter/step	*Element*										
	As	*Cd*	*Cr*	*Cu*	*Fe*	*Mn*	*Ni*	*Pb*	*Se*	*Tl*	*Zn*
Instrumental parameters											
Instrument[a]	4100	5000	4100	5000	4100	4100	5000	5000	4100	4100	5000
Wavelength, nm	193.7	228.8	357.9	324.8	248.3	279.5	232.0	283.3	196.0	276.8	213.9
Slit, nm	0.7	0.7	0.7	0.7	0.2	0.2	0.2	0.7	2.0	0.7	0.7
Source, mA, W	EDL	EDL	HCL	HCL	HCL	HCL	HCL	EDL	EDL	EDL	HCL
	8	5	25	15	30	20	20	8	6	7	10
Temperature programme											
Drying	all elements 120°C, ramp 1 s, hold 50 s, for all elements										
Charring	ramp 10 s, hold 20 s										
Temperature, °C	1200	800	1500	1000	1400	1300	1100	850	1100	1000	700
Atomization	max. power heating, hold 5 s, for all elements										
Temperature, °C	2300	1600	2300	2000	2100	1900	2300	1600	1900	1800	1800
Internal gas flow, ml/min[b]	0	0	0	0	0	0	0	0	0	0	300
Cleaning	2400°C for Zeeman 4100 ZL, 2700°C for Zeeman 5000										
Cooling	20°C, hold 10 s, for all elements										
Modifier[c]	A	B	C	A	C	C	no	B	A	A	C

[a] 4100: Spectrometer system Zeeman-AAS 4100 ZL; 5000: Zeeman-AAS Model 5000.

[b] Internal gas flow (Ar) of 250 ml/min (AAS-4100 ZL) and 300 ml/min (AAS 5000), respectively; gas stop in atomization step.

[c] A: 32.5 μg $Pd(NO_3)_2$ + 10 μg $Mg(NO_3)_2$; B: 253 μg NaH_2PO_4 + 10 μg $Mg(NO_3)_2$; C: 50 μg Mg $(NO_3)_2$.

After cooling, the sample solutions were transferred into 10-ml quartz flasks and filled up with doubly-distilled water. 20μl portions of these solutions were taken for analysis by AAS using operating parameters and temperature programme given in Tables 1 and 2. The addition method was used for standardization. Peak areas were recorded. The standard reference material BCR 1575 'Pine Needles' was processed in the same way.

Table 2 Instrumental parameters for flame AAS (three-slot burner, air-acetylene oxidizing flame).

Parameter	*Element*	
	K	*Na*
Wavelength (nm)	766.5	589.0
Slit (nm)	0.7	0.7
Radiation source	HCl	HCl
Setting (mA)	6	8

INAA

For determinations via short- and medium-lived indicator radionuclides (^{76}As, ^{82}Br, ^{42}K, ^{56}Mn, ^{24}Na), sample portions of about 30–50 mg of dry tobacco, smoke condensate, the standard reference material (BCR-1575, Pine Needles), the internal standard material Nicotinic Acid and multi-element standards containing appropriate concentrations of each element to be determined were irradiated simultaneously in polyethylene capsules (*Stichting Hart Wool Research Foundation*, Amsterdam, the Netherlands) for 2 h at a thermal neutron flux of 1.3×10^{13} cm^{-2} s^{-1} (ratio of flux of thermal and fast neutrons, 7:1) in the FRM-1 reactor (Garching, Germany). For the determination of the other elements via long-lived indicator radionuclides, samples and multielement standards were irradiated in Suprasil® quartz ampoules (Heraeus, Hanau, Germany) for 3 days at a thermal flux of 1×10^{14} cm^{-2} s^{-1} (ratio of flux of thermal and fast neutrons, 14:1) in the FRG-2 reactor (Geesthacht, Germany). Before use, the polyethylene capsules were washed with HNO_3, the quartz ampoules were etched with HNO_3 (65%) and afterwards with HF (40%) selectipur (Merck). Multi-element standards were prepared under laminar-horizontal flow clean-bench conditions by pipetting suitable volumes of the respective element stock solutions into the ampoules and drying in a desiccator over P_2O_5 at reduced pressure and room temperature. Before counting, the surface of the polyethylene ampoules was cleaned by washing with subboiled HNO_3 whereas the Suprasil® quartz tubes were etched with HF (40%). For controlling the blank, empty ampoules were irradiated simultaneously and processed in the same way. Counting was performed by using the high resolution γ-ray spectrometer system described above. The most important nuclear data for the indicator radionuclides are given in Table 3.

RESULTS AND DISCUSSION

For our investigations, we selected the cigarette brand with the highest market share in Germany, Marlboro, and, for comparison, the average cigarette of the German market, C-20.

Three methods of collecting the total particulate matter of the cigarette smoke have already been described in the literature: the collection in cold- or solvent-traps[10–19], on cambridge-filters[20–31], and in an electric field[2, 32–35,37].

Our preliminary experiments with different cooled solvent traps filled with acetone, methanol, n-hexane, chloroform, diluted nitric acid and different mixtures of these solvents showed that the collection of the smoke was not quantitative. Because of the low concentration levels of the investigated elements in the smoke, it is necessary to collect smoke condensate of at least 20 cigarettes in order to obtain the sample and determination portions required for analysis by INAA and AAS. However, after smoking of two cigarettes, the traps got dogged by precipitated particles of smoke condensate.

In another experiment, the smoke condensate was collected on a cambridge-filter. Also in this case smoke condensate of only two cigarettes could be collected. The unsatisfactory efficiency and the high blanks of the filters do not recommend the filtration sampling methods for this purpose.

Compared with the two sampling methods mentioned above, electrostatic precipitation of smoke condensate proved to be a well suited sampling method offering several advan-

Table 3 Nuclear data for the indicator radionuclides (IRN) used in NAA [52,53].

Element	*IRN*	*Isotopic abundance, %*	$t_{1/2}$	*main γ-line, keV (and intensity, %)*	
Ag	^{110m}Ag	48.17	250.4 d	657.7	(94.4)
As	^{76}As	100	26,32 h	559.1	(45.0)
Au	^{198}Au	100	2,695 d	411.8	(95.5)
Br	^{82}Br	49.3	1.47 d	776.5	(83.5)
				554.3	(70.7)
				619.1	(43.2)
Cd	^{115}Cd	28.72	53.5 h	527.9	(27.5)
	^{115}In			336.3	(45)
Ce	^{141}Ce	88.48	32.51 d	145.5	(48)
Co	^{60}Co	100	5.272 a	1173.2	(99.9)
				1332.5	(100)
Cr	^{51}Cr	4.35	27.71 d	320.1	(9.8)
Fe	^{59}Fe	0.3	44.6 d	1099.3	(56)
				1291.6	(44)
Hg	^{203}Hg	29.7	46.6 d	279.2	(81.5)
K	^{42}K	6.7	0.51 d	1524.7	(17.9)
Mn	^{56}Mn	100	0.107 d	846.8	(98.9)
Na	^{24}Na	100	0.63 d	1368.5	(100)
Ni	^{58}Co	67.88	70.78 d	810.6	(99.4)
Sb	^{122}Sb	57.25	2.72 d	564.1	(63)
	^{124}Sb	42.75	60.3 d	602.7	(98.1)
				1691	(50)
Sc	^{46}Sc	100	83.8 d	889.3	(100)
Se	^{75}Se	0.9	120 d	135.9	(58)
				264.5	(58.5)
Th	^{233}Pa	100	27 d	311.9	(38)
U	^{239}Np	99.27	2.35 d	106	(21)
				277.6	(12)
Zn	^{65}Zn	48.9	243.7 d	1115.5	(49.8)

tages. The blanks can be minimized by making all parts of the smoking machine and the electrostatic trap which can have contact with smoke or smoke condensate of high purity materials such as PTFE and quartz. For this reason, we replaced the original nickelbrass electrode by one made of high-purity aluminium. Aluminium was not included in this study and a possible contamination of smoke condensate with this element does not give rise to any interferences in the determination of the analyte elements by AAS or INAA. Another advantage of the electrostatic precipitation trap is that it enables to collect smoke condensate of 100 cigarettes in the quartz tube. From the average cigarette weight of 0.96 ± 0.02 g, it follows that the concentration values expressed in ng/cig. are almost equal with values expressed in ng g^{-1} (1 ng/cig. = 1.04 ng g^{-1}). The effectivity of the collection rate of the total particulate matter of the smoke was controlled by a Cambridge filter placed at the end of the outlet pipe of the smoking machine. It was reported[33,38] that some elements, especially nickel, can appear in the gas phase of the cigarette smoke, too. Therefore, in order to check the possible occurrence of this effect, the gas phase of the smoke was trapped in conc. HNO$_3$ in two quartz flasks connected in sequence to the outlet pipe of the smoking machine. The concentration of nickel determined in nitric acid was below the limit of detection corresponding to 0.5 ng/cig. when sampling the condensate of 200 cigarettes. In the condensate of the two investigated cigarette brands, nickel contents of 1.1 and 1.5 ng/cig. were determined.

The element concentrations determined by both methods in tobacco and smoke condensate of both cigarette brands are given in Tables 4 and 5, respectively, together with the achievable limits of detection (obtained by applying the criterion of three standard deviations of the fluctuation of the blank (AAS) or of the Compton background (INAA)). The concentrations determined in the standard reference material "Pine Needles" and the nicotinic acid are given in Table 6. As can be seen from Tables 4 and 5, the results of the two analytical methods agree within the experimental uncertainties in all comparable cases. The mean values of the two methods differ by more than 20% only in 15 of 85 comparable cases.

In addition to the analytical errors, the uncertainties include possible inhomogeneities regarding the distribution of the elements in tobacco and smoke condensate. As is evident from the low blanks and their fluctuation, occurrence of contamination errors was of low relevance.

Table 4 Concentrations of elements determined in tobacco of the cigarettes C-20 and Marlboro and limits of detection.

Element	*Concentration (μg/g)*				*Limit of detection (μg/g)*	
	C-20		*Marlboro*			
	INAA	*AAS*	*INAA*	*AAS*		
Ag	<0.2	-	<0.02	-	<0.02	(INAA)
As	<0.4	0.41 ± 0.02	<0.4	0.29 ± 0.04	<0.25	(AAS)
Au	<0.0003	-	<0.006	-	<0.0003	(INAA)
Br	110.8 ± 5.2	-	84.8 ± 4.9	-	<5.0	(INAA)
Cd	<2.4	1.95 ± 0.21	<2.5	1.81 ± 0.16	<0.004	(AAS)
Ce	2.0 ± 0.1	-	2.0 ± 0.2	-	<0.02	(INAA)
Co	0.57 ± 0.10	-	0.76 ± 0.02	-	<0.0005	(INAA)
Cr	1.2 ± 0.2	1.1 ± 0.2	1.7 ± 0.3	1.3 ± 0.3	<0.02	(INAA)
Cu	-	9.70 ± 0.42	-	6.90 ± 0.72	<0.08	(AAS)
Fe	581 ± 22	-	478 ± 10	-	<0.03	(AAS)
Hg	<0.09	-	<0.06	-	<0.06	(INAA)
K	32000 ± 1600	32500 ± 2500	31600 ± 2100	35000 ± 2000	<0.2	(INAA)
Mn	166.9 ± 18.9	135.3 ± 10.5	180.5 ± 28.6	110.1 ± 39.8	<0.02	(INAA)
Na	348 ± 76	446 ± 88	356 ± 56	483 ± 120	<0.5	(INAA)
Ni	2.1 ± 0.2	2.4 ± 0.3	2.4 ± 0.1	2.3 ± 0.1	<0.08	(AAS)
Pb	-	1.3 ± 0.9	-	0.77 ± 0.05	<0.07	(AAS)
Sb	0.07 ± 0.02	-	0.06 ± 0.01	-	<0.0005	(INAA)
Sc	0.20 ± 0.04	-	0.20 ± 0.05	-	<0.0002	(INAA)
Se	0.18 ± 0.05	-	0.20 ± 0.05	-	<0.1	(INAA)
Th	<0.2	-	<0.2	-	<0.2	(INAA)
Tl	-	<1.0	-	<1.0	<1.0	(ASS)
U	<0.4	-	<0.4	-	<0.4	(INAA)
Zn	35.8 ± 7.0	49.8 ± 2.6	37.3 ± 6.6	49.0 ± 5.9	<0.2	(INAA)

Table 5 Concentrations of elements in smoke condensate of the cigarettes C-20 and Marlboro and limits of detection.

Element	*Concentration (ng/cig.)*[a]				*Limit of detection (ng/cig.)*	
	C-20		*Marlboro*			
	INAA	*AAS*	*INAA*	*AAS*		
Ag	<0.2	-	<0.3	-	<0.2	(INAA)
As	4.7 ± 0.3	5.3 ± 0.3	4.1 ± 0.4	4.1 ± 0.4	<0.5	(AAS)
Au	<0.03	-	<0.03	-	<0.03	(INAA)
Br	340 ± 16	-	380 ± 40	-	<0.5	(INAA)
Cd	55 ± 8	56 ± 3	64 ± 3	65 ± 3	<0.06	(AAS)
Ce	<0.3	-	<0.3	-	<0.3	(INAA)
Co	0.13 ± 0.02	-	0.08 ± 0.03	-	<0.005	(INAA)
Cr	<2.6	<1.7	<0.6	<2.2	<0.6	(INAA)
Cu	-	13.3 ± 1.3	-	13.0 ± 0.6	<2.0	(AAS)
Fe	<28	13.9 ± 5.5	<22	16.8 ± 0.3	<0.5	(AAS)
Hg	<1.0	-	<0.5	-	<0.5	(INAA)
K	39000 ± 1700	39000 ± 3000	47000 ± 3300	44500 ± 5700	<3.0	(INAA)
Mn	2.1 ± 0.3	2.8 ± 0.5	2.2 ± 0.1	2.6 ± 0.5	<0.1	(INAA)
Na	1200 ± 190	1550 ± 170	2270 ± 260	1760 ± 250	<5.0	(INAA)
Ni	<3.0	1.10 ± 0.98	<3.0	1.46 ± 0.98	<1.0	(AAS)
Pb	-	40.0 ± 1.8	-	32.0 ± 1.4	<1.0	(AAS)
Sb	0.10 ± 0.01	-	0.13 ± 0.01	-	<0.005	(INAA)
Sc	<0.0005	-	<0.0018	-	<0.0005	(INAA)
Se	1.8 ± 0.1	-	1.5 ± 0.2	-	<0.07	(INAA)
Th	<0.010	-	<0.010	-	<0.010	(INAA)
Tl	-	<9.8	-	<11.3	<10.0	(AAS)
U	<2.0	-	<3.0	-	<2.0	(INAA)
Zn	70.0 ± 20	91.5 ± 15.2	134.7 ± 10.8	127.3 ± 27.5	<0.4	(INAA)

[a] 1 ng/cig. corresponds in average 1.04 ng per gramm of TSC.

Table 6 Results obtained for the standard reference material 'Pine Needles' (BCR 1575) and for the internal standard 'Nicotinic Acid'.

Element	*Concentration (μg/g)*			
	Pine Needles		*Nicotinic Acid*	
	determined value	*certified value*	*determined value*	*certified value*
As	-	-	1.5 ± 0.1	1.5 ± 0.2
Cd	-	-	2.1 ± 0.1	2.1 ± 0.2
Co	-	-	10.0 ± 0.1	8.5 - 11.4
Cr	-	-	2.7 ± 0.2	3.0 ± 0.1
Cu	3.0 ± 0.4	3.0 ± 0.3	-	-
Mn	680 ± 83	675 ± 15	-	-
Ni	-	-	3.2 ± 0.1	3.1 ± 0.3
Pb	9.8 ± 0.4	10.8 ± 0.5	-	-
Sb	0.08 ± 0.03	(0.2)*	-	-
Sc	0.042 ± 0.001	(0.03)*		- -
Zn	-	-	2.1 ± 0.2	2.0 - 3.2

* Non-certified values.

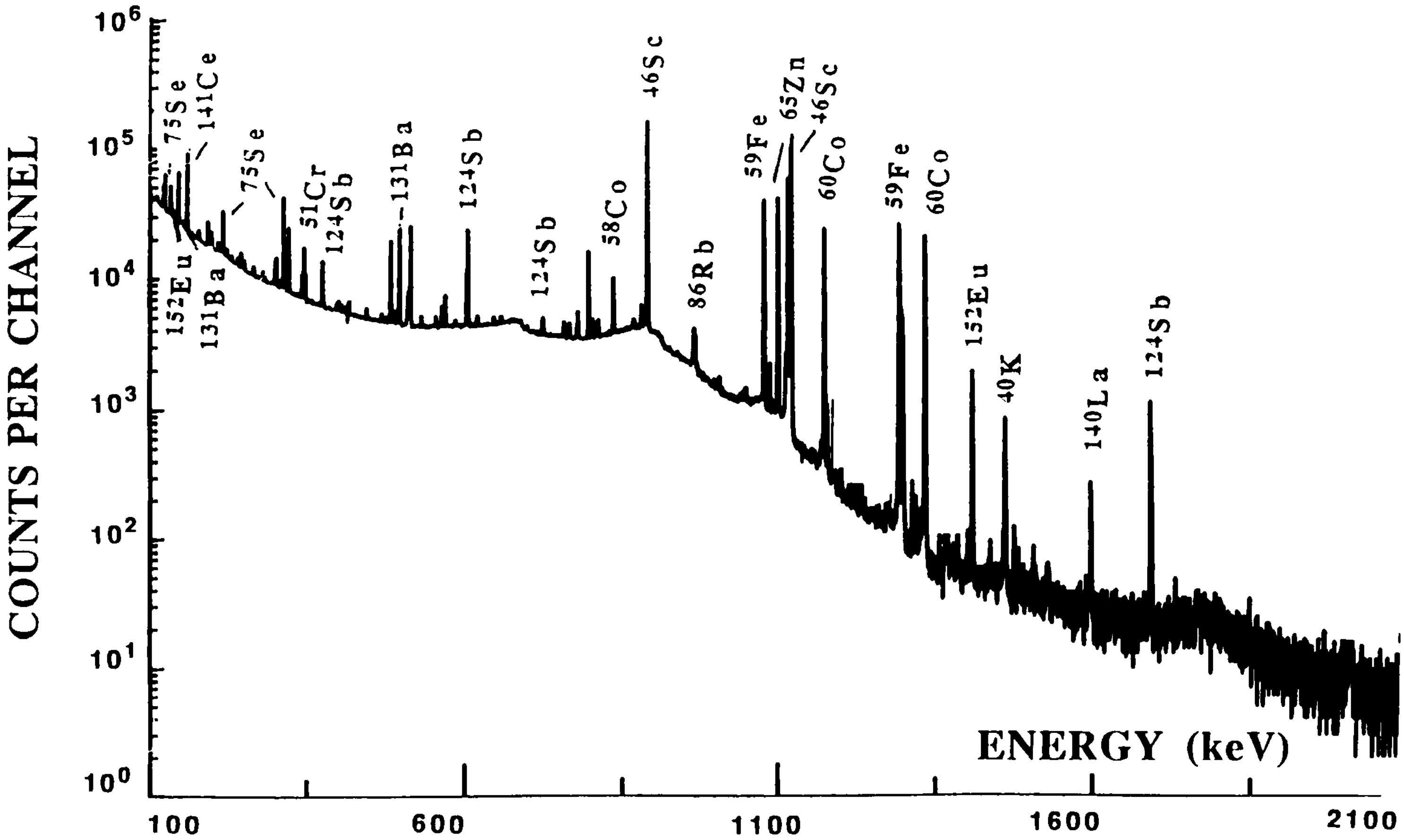

Figure 2 Gamma spectrum of a neutron-irradiated cigarette smoke condensate (~100mg); Øth = 10^{16} cm^{-2} s^{-1}; t_{irr}=3d; t_d = 27d; t_c = 60,000s.

GFAAS using the STPF concept enables a quick and reliable determination of a number of toxic elements in tobacco and smoke condensate including As, Cd, Cr (only in tobacco), Cu, Fe, Mn, Ni, Pb and Zn. However, the favourable performance parameters of these analytical methods with respect to detection limits and reliability are, to a considerable degree, the result of using the advantageous pressure microwave decomposition of the samples. Owing to utilization of quartz vessels and small volumes of subboiled nitric acid, extremely low blank levels can be achieved with this rapid decomposition procedure, resulting in excellent limits of detection also for elements with relatively high overall concentrations. To avoid blanks in homogenizing the tobacco, the samples were pulverized in a PTFE capsule in the microdismembrator. Because of high detection sensitivity and the complete absence of reagent blank, neutron activation analysis is a technique well suited for the analysis of both materials investigated, but especially for analysis of the smoke condensate. Since no dominant activity is produced during irradiation of these two materials with reactor neutrons, extremely low limits of detection for a large number of elements can be achieved even with the direct purely instrumental performance of the NAA. As an example, Figure 2 shows a γ-ray spectrum of long-lived indicator radionuclides of a neutron-activated smoke condensate sample.

In Table 7, our results are compared with those published in the literature. In most cases, our results are in the lower region of the published data or they are even lower than all published values (As, Co, Cr, Cu, Mn, Ni, Sb, Sc, Zn). The reason for the large scatter of the concentrations of the individual elements determined in smoke of the main stream are manifold. One reason is the very different levels of the elements in the various tobacco types investigated. Another one is connected with the development of trace-element analysis and the associated difficulties to control the systematic errors in the analytical process. Especially in older investigations of the low-concentration-level elements, the high results obtained are probably due to contaminants introduced during sampling and/or analysis from the working atmosphere, solvents, reagents and vessels.

During smoking a cigarette, the elements contained in tobacco are to a certain percentage transferred into main and side stream and the rest is retained in the filter and the ash (see Fig. 1). From the analytical results obtained we estimated the per cent transfer of the elements from tobacco into the smoke of the main stream which are given in Table 8. The condensate of one C-20 cigarette weighing 960 mg amounts to 13 mg, i.e., 1.35% (all average data). From this data it is evident that as a result of an enrichment process the heavy metal elements with per cent transfers of more than 1.35%, cadmium and lead, are present at higher concentrations in the smoke than in tobacco. On the other hand, for the elements Ce, Co, Fe, Mn and Ni, the transfer is extremely low (<0.05%), i.e., these elements are mainly retained in ash and filter. Surprisingly, arsenic and antimony show much lower relative transfer factors than would be expected on the bases of their volatilities at temperatures realized during smoking. Their per cent transfers are lower than those for cadmium and lead, and are near to the percentage of smoke condensate production.

As a result of the contents in tobacco and the transfer factors, the elements are present in the smoke at very different concentrations. As is evident from Table 4, the elements determined in tobacco smoke condensate can be assigned to four groups by concentration level: elements at sub—ng/cig. level (Ag, Au, Ce, Co, Cr, Hg, Sb, Se, Th and U), at low-ng/cig. level (As, Mn, Ni and Se), at medium- and upper-ng/cig. level (Br, Cd, Cu, Fe, Pb

Table 7 Comparison of element contents in cigarette smoke with data reported in the literature.

Element	*Content, ng/cig.*			
	Our results (C-20/Marlboro)	*Reported data*	*Ref.*	*Collection mode**
As	5.0/4.1	10	10	a
		50	21	a
Cd	55.2/64.2	30	10	a
		200	21	a
		7–210	11	a,b
		100	23	b
		147.4	24	b
		178	33	a,b,c
		30–80	2	c
		220–270	37	a,b,c
Co	0.13/0.08	10	10	a
		11	13	a,b
		12	36	c
Cr	<1.7/<0.6	4.8	36	c
Cu	13.3/13.0	20	36	c
		100	28	a,b
Mn	2.5/2.4	40	28	a,b
		60	33	a,b,c
Na	1375/2017	220	13	a,b
Ni	1.10/1.46	4	16	c
		5	10	a
		5–67.5	11	a,b
		72.6	24	b
		100	23	b
		225	33	a,b,c
Pb	40.0/32.0	34	16	c
		50	10	a
		100	17,21,37	a,b,c
		65–610	11	a,b
Sb	0.10/0.13	10	21	a
Sc	<0.0005/<0.0018	0.027	13	a,b
Se	1.8/1.5	20	21	a
Zn	80.0/131.0	248	16	c
		600	28	a,b

*) collection mode: a, cold-or solvent trap; b, Cambridge filter; c, electrostatic trap.

and Zn) and at μg/cig. level (K and Na). On the basis of their toxicities and concentrations in the tobacco smoke, several elements have to be considered as possibly increasing the health hazard potential for smokers, particularly in terms of chronic lung diseases[5,6,7]. The most relevant elements appear to be cadmium and lead. An uptake of cadmium (biological half-life, 20 years) via the respiratory tract causes a 10-fold higher resorption than an uptake via food. The estimated average uptake of this element through food is about 15 mg per day resulting in a resorption of 0.75 mg[5]. Smoking 20 cigarettes daily can lead to a resorption

Table 8 Per cent transfer of elements from tobacco into smoke compared with data reported in literature.

Element	*Transfer(%)*			
	C-20	*Marlboro*	*Published values*	*Ref.*
As	1.25	1.46	0.5–6.3	15,17,
Br	0.32	0.47	0.02-2.4	13,15,20
Cd	2.95	3.68	5.1/4.65	2,10
Ce	<0.02	<0.016	-	-
Co	0.024	0.011	0.5/0.87	15,20
Cr	<0.15	<0.04	1.74/0.43	15,20
Cu	0.14	0.20	0.4–1.7	17
Fe	0.0025	0.0037	0.05–0.17	15,17,20
K	0.12	0.14	0.2-0.5	13,17
Mn	0.002	0.002	0.003-0.006	13
Na	0.36	0.50	0.3-1.0	17
Ni	0.05	0.06	1.79	16
Pb	3.20	4.32	3.9-4.8	17
Sb	0.14	0.22	1.9	20
Se	1.06	0.79	2.5	15,20
Zn	0.20	0.32	0.2-2.3	15,16,17,20

of 1.5 mg/day, causing an increase of the cadmium content in the lung tissues of smokers by a factor of 2 relative to non-smokers[5,6,7].

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