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## Accurate measurement of main aerosol constituents from heated tobacco products (HTPs): Implications for a fundamentally different aerosol



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### ABSTRACT

ISO 4387 Standard determines the main aerosol constituents (total particulate matter, water, nicotine, and nicotine-free-dry-particulate matter, referred to as “tar”) in cigarette mainstream smoke (ISO, 2000). Heated Tobacco Products (also called Tobacco Heating Products or Heat-not-Burn Products) are designed to form aerosol by heating tobacco rather than burning like in combustible cigarettes. In this study we have evaluated the suitability of ISO 4387 Standard to be adapted for quantifying main aerosol constituents for HTP aerosol. HTP emissions have much higher levels of water and humectants (e.g., glycerol) in dynamic equilibria between gaseous and particulate phases. Several modifications to ISO 4387 Standard on aerosol collection were tested to improve the accuracy and reliability of aerosol capturing, with minimal deviation to the standard method. The proposed modifications are readily adoptable by laboratories already practicing the Standard for cigarette smoke analyses. Taking collectively with other available aerosol chemistry and biological results on HTPs in the literature, they show a fundamentally different aerosol in HTPs and call for category-specific product standards and terminology.

### 1. Introduction

Electrically heated tobacco products (HTPs, or eHTPs) are designed to deliver tobacco volatiles and nicotine aerosol in a markedly different way to cigarette smoke. Regulatory laboratories have begun to test these products (RIVM, 2018; Mallock et al., 2018; Li et al., 2018). For example, a recent study funded by the German Federal Institute for Risk Assessment (BfR) showed that in addition to significantly reduced toxicant levels in HTP emissions, due to high levels of humectant and water found in the nicotine-free-dry-particulate-matter (NFDPM), direct comparison of these values with those from combustible cigarettes would be misleading (Mallock et al., 2018).

Cigarette smoke is a complex aerosol system with over 7000 identified chemicals present as gases, and condensed phase compounds (Rodgman and Perfetti, 2013). Basic characterisation of cigarette smoke starts with gravimetric quantification of its total particulate matter, and the main aerosol constituents, including nicotine and water, when smoked under a standardised smoking regime (Borgerding and Klus, 2005). Over the years the International Organization for Standardization (ISO) Methods ISO 4387 (ISO, 2000), ISO 3308 (ISO, 2012), ISO

10362-1 (ISO, 1999) and ISO 10315 (ISO, 2013) have set the requirements for preparation of cigarettes, machine-smoking and smoke collection under a set of defined parameters for analytical purposes. The capture of smoke through a glass fibre filter pad called a Cambridge filter pad (CFP) is one of the most important procedures. CFPs have a greater than 99.9% trapping efficiency for smoke aerosol for particles of 0.3 µm diameter and larger under the standard machine puffing conditions of ISO 3308 (ISO, 2012), a key requirement for the task. The trapped smoke matter is measured gravimetrically and corrected for water and nicotine content. The World Health Organization (WHO), through its Tobacco Free Initiative network of laboratories (TobLabNet) also has a list of recommended test methods on cigarette smoke analyses ([http://www.who.int/tobacco/global\\_interaction/toblabnet/en/](http://www.who.int/tobacco/global_interaction/toblabnet/en/)).

Total particulate matter (TPM) is defined by ISO 4387 as the condensed-phase portion of the mainstream smoke which is trapped by the CFP, expressed as milligrams per cigarette. Dry particulate matter (DPM) is defined as the total particulate matter after mathematical deduction of its water content. Nicotine-free dry particulate matter (NFDPM) is defined as the dry particulate matter after deduction of its nicotine content. NFDPM values have commonly been referred to as

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cigarette “tar”, which is still printed on cigarette packs in some jurisdictions, together with nicotine and carbon monoxide levels. It should be noted that the particulate water content is not an account of the total smoke water produced by a lit cigarette. A small portion of gaseous water (steam) will pass through the CFP pad and not be measured by the technique. Standard procedures in ISO 4387 are designed to address any variabilities likely to be caused by trapping of this gaseous fraction of water.

Dynamic changes (sometimes called ageing effects) in physical (equilibria between gaseous, semi-volatile and condensed phases) and chemical (e.g., free radical reactions) properties, post formation, are inherent to cigarette smoke and HTP aerosol alike. Current commercial electrical HTPs operate by heating a section of tobacco to around 200–350 °C to generate an aerosol (Eaton et al., 2018; Schaller et al., 2016a, b). The tobacco rods are enclosed within an electronic device during operation and the heat supplied from the device evaporates and distills volatile and semi-volatile constituents from the tobacco, together with added agents such as glycerol, to form the aerosol. This contrasts markedly to the tobacco thermophysics in a lit cigarette, in which tobacco is exposed to a range of temperatures up to above 900 °C (Baker, 1974). Another key difference is that in an HTP, it is the same portion of the tobacco that is repeatedly heated to form aerosol, rather than a new tobacco section feeding into the burning process, by consuming a lit cigarette rod. Other differences in thermophysical and thermochemical processes between the two systems also exist (see later). The most noticeable effect for HTP aerosol collection is the large quantity and higher temperature of water/steam in the aerosol steam, leading to larger variability during machine-puffing and aerosol recovery stages.

In fact, evidence of this challenge has been seen in cigarette smoke testing. For example, a technical report by the ISO discusses these issues when examining systematic differences in cigarette smoke yields when switched to use intense puffing regimes (ISO/TR, 2015). Difficulties in the gravimetric determination of TPM and NFDPM are noted when water becomes a major component of smoke condensate, in this case due to more intense smoking and higher intensity of burning tobacco. There are a number of parallels and also differences between the intense smoking of cigarettes and the way HTPs emit aerosol. Chiefly HTPs give minimal pyrolysis of tobacco and distil water all through mainstream. HTP tobacco stick design incorporates water vapour and aerosol temperature management as its main task, rather than particulate filtration as in cigarettes (Eaton et al., 2018; Schaller et al., 2016a, b). The sensory perception of initial puffs is substantially warmer for HTPs. These factors influence the water condensation and aerosol formation process for HTPs, and present questions for the usability of the ISO 4387 Standard for effective and accurate collection of HTP aerosol. Previously, Borgerding et al. (1990) have reported capricious water measurements in a charcoal-tipped heated tobacco

product. They proposed an in-holder rinsing method that appeared to recover an additional component of gravimetrically recorded water but they described this technique as being cumbersome. The incorrect accounting for water by following the ISO 4387 standard method led to a reported approximately 25% higher calculated NFDPM for their product. More recently, Ghosh and Jeannet (2014) reported that the ISO standard methods are not appropriate for water determination for HTP emissions and proposed additional equipment for trapping and extracting the water. Using this *in-situ* extraction approach, they reported gravimetric water yields for a commercial HTP (THS 2.2) to be 44.65 mg/tobacco rod (SD = 2.47), in contrast with a water yield of 37.55 mg/tobacco rod (SD = 2.81) for the same product when following the ISO 4387 without modification. The method they proposed however requires custom-made components, which are said to be “neither standardized nor applicable for surveillance authorities” (Mallock et al., 2018).

This challenge has been reflected in the wider reported values for these products in the literature. For THS 2.2, the water level has been variously reported as  $25.4 \pm 2.0$  mg/tobacco rod (Forster et al., 2018) using the ISO 4387 method,  $36.5 \pm 1$  mg/tobacco rod with a similar product and citing standard ISO testing methodology for NFDPM assessment (Gonzalez-Suarez et al., 2016),  $36.5 \pm 3.1$  mg/tobacco rod,  $39.4 \text{ mg} \pm 3.1$  mg/tobacco rod, citing the modification due to Ghosh and Jeannet (Schaller et al., 2016a), and  $32.1 \pm 6.5$  mg/tobacco rod (Schaller et al., 2016b) following a standard combustible method. Consequently the breadth of reported values highlights a large uncertainty with reported NFDPM values, potentially leading to erroneous reporting to regulators and miscommunication to the consumer.

Our aim for this study was to evaluate a set of critical parameters on aerosol capturing as set in the ISO 4387 Standard, taking into the account the differences in aerosol formation and aerosol properties between combustible cigarettes and HTPs. As far as possible, we proposed practical modifications and evaluated their effects on aerosol capturing, which could be considered as interim measures to improve the ISO Standard’s consistency for wider laboratory assessment of HTPs, until HTP category-specific standards are evaluated and set by international standard agencies.

## 2. Experimental

Experimental work described here was performed by a contract laboratory (Labstat International ULC, Kitchener, ON, Canada). As far as possible, the experimental procedures followed the conditions stipulated by ISO 4387 (ISO, 2000) and assessed simple modifications targeting those steps thought to affect the main HTP aerosol collections.

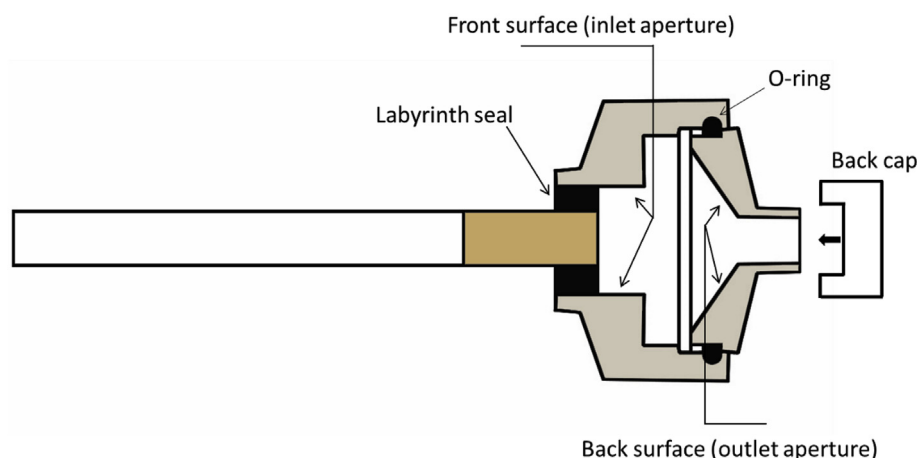


Fig. 1. A schematic drawing of a Cambridge filter pad assembly, connected to a test cigarette.

## 2.1. Key parameters in smoke and HTP aerosol capturing

The standard method for collection of total particulate matter uses a CFP hosted in a standardised pad holder (Fig. 1). The material (composition, surface finish, etc) of the pad holder and the internal cavity volume are factors that may affect the degree of water condensation when passing an aerosol stream. Most evaporative loss may occur when the pad holder is opened along the o-ring joint, and the CFP is taken out for weighing. This procedure has been evaluated and accepted for cigarette smoke analysis.

The measurement of TPM, nicotine and water (to derive NFDPM) begins with the weighing of the CFP assembly before and after the machine puffing run. Once the pad holder (the entire CFP pad assembly without the front and back caps) is opened, the CFP is removed and placed in a solvent (isopropyl alcohol), and then the inside front surface of the filter pad holder and the apertures are wiped with two quarters of a clean CFP which are also placed in the solvent. This procedure assumes negligible evaporation/condensation in the time between the end of smoking/puffing, opening the holder and placing the CFP in the solvent. It also assumes a full recovery of condensed material by wiping the front internal surface of the pad holder and no deposition collecting on the rear surface of the holder or in the inlet aperture and labyrinth seals (Fig. 1). These assumptions and parameters were varied during the evaluation of HTP aerosol collection (see later). The standard plastic CFP pad holders and caps were purchased from Cerulean (Rockingham Drive, Linford Wood East, Milton Keynes, MK14 6LY, UK).

A full list of all the operation parameters investigated by this study is given in Table 1. Water and nicotine yields were measured using ISO 10362-1 (ISO, 1999) and ISO 10315 Standards (ISO, 2013) respectively, TPM was recorded gravimetrically and NFDPM was formally calculated for each product variant. Each method was also accompanied by an air blank. For the tests using impingers, the air-blank results were used to correct for background moisture from laboratory air. Video recordings of the extraction procedures were made to check the consistency of operator's procedures and were used to improve staff training (not presented here).

## 2.2. Machine puffing and conditioning parameters

Two commercial electrical HTPs were used in this study. For tobacco samples, the overwrap was removed and the packs stored closed

**Table 1**  
Summary of the method parameters investigated.

Test	Article	Reps	Rods per replicate	Puff Count	Puffing	Modification
1.1	THS 2.2	5	3	12	HCI	Standard method with no modifications
1.2	THS 2.2	5	3	12	HCI	Modified by additional wiping of the front and back of the Pad holder using one additional quarter pad. A second additional quarter pad was used to wipe the inlet aperture assembly. Both were extracted with 10 mL of extraction solvent and reported separately.
1.3	THS 2.2	5	3	12	HCI	Modified by adding a secondary impinger with extra coarse frit containing 20 mL extraction solvent, in series behind the Pad holder. The impinger result (including extraction/rinsing of the tubing) was reported separately.
1.4	THS 2.2	5	3	12	HCI	Modified by eliminating the CFP and using an impinger with an extra coarse frit. Included clearing puffs and extraction/rinsing of the tubing.
2.1	THS 2.2	5	3	12	HCI	Standard method with no modifications
2.2	THS 2.2	5	3	12	HCI	Modified by extra care with capping plus additional wiping of front and back of holder, inlet aperture and labyrinth seals
3.1	THS 2.2 THP 1.0M THP 1.0T	17	3	12 8 8	HCI	Standard method with no modifications
3.2	THS 2.2 THP 1.0M THP 1.0T	17	3	12 8 8	HCI	Modified by combined extraction of CFP, wiped inner surface of holder using additional 2 X 1/4 pad, plus wiped inner surface of front aperture of Pad holder (termed inlet aperture) which is the normal position of neoprene washer that is not used, using a 1/4 pad, plus wiped inner surface of front and back aperture covers for the Pad holder using a 1/4 pad. Main pad and all pad segments were extracted together.
3.3	THS 2.2	17	3	12	HCI	As modification 3.2 but additional pad segments were extracted and reported separately
3.4	THP 1.0M THP 1.0T	5	5	4	ISO	Standard method with no modification

**Table 2**

Two electrically heated tobacco products used in the study.

Sample Code	Description	Status
THS 2.2	IQOS Essence Heatstick	Commercial product
THP1.0(M)	glo Intensely Fresh Neostik	Commercial product
THP1.0(T)	glo Bright Tobacco Neostik	Commercial product

at conditions consistent with ISO 3402 (ISO, 1999) for a minimum of 48 h and a maximum of 120 h. Due to the high level of glycerol in HTP tobacco products, the tobacco sticks are more susceptible to environmental moisture uptake. In the absence of a standard conditioning protocol, conditioning by removing overwrap was adopted. CFPs used for the study were equilibrated under the same conditions. The mainstream aerosol was generated on a Borgwaldt linear smoking machine type LM20X (Borgwaldt KC GmbH, Hamburg, Germany). Puffing parameters followed either the HCI (Health Canada, 2000) or the ISO 3308 (ISO, 2012) as applicable. In brief these are 35 mL puff volume, 2 s puff duration, once every 60 s with a bell-shaped puff flow for ISO puffing, or 55 mL puff volume, 2 s puff duration, once every 30 s with a bell-shaped puff flow for HCI puffing. Ventilation blocking was not used for the HTPs. In all cases the neoprene washer was omitted from the inlet aperture of the pad holder assembly.

## 2.3. Other materials

The two commercial HTPs and their tobacco consumable variances are listed in Table 2. THS 2.2 comprised the IQOS™ heating device with Essence Heatstick™ which were purchased from a retail outlet in Japan. The THP1.0 comprised the glo™ heating device, with the variant THP1.0(T) being Bright Tobacco Kent Neostiks™, and THP1.0(M) being Intensely Fresh Kent Neostiks™, both sourced from Japan.

## 2.4. Data analysis

The mean results are tabulated along with replicate numbers and their standard deviations. Key results are also presented as bar charts with error bars of 1 standard error, to allow a visual assessment of significance of differences in the mean values. Where relevant, direct comparisons between procedures are made using student's two-sample *t*-test with Welch's correction. The following procedure was adopted to

**Table 3**  
Mainstream water yields in THS 2.2 emissions during Phase 1 (n = 5). Values in bracket are percentage of water in each component.

Method	Part analysed	Mean (mg/tobacco rod)	SD	Total Water (mg/tobacco rod), by method	SD
1.1 Standard method	Standard method	24.7 (100)	1.6	24.7	1.6
1.2 Additional wiping/extraction of holder, inlet aperture	Pad	20.5 (57.1)	1.7	35.9	1.9
	Holder	13.7 (38.2)	0.4		
	Inlet aperture	1.7 (4.7)	0.4		
1.3 Addition of impinger	Standard method	26.4 (71.4)	1.3	33.1	2.2
	Coarse fritted Impinger	8.6 (23.2)	1.2		
	Air blank from impinger	2.0 (5.4)	1.3		
	Impinger	38.4 (94.1)	5.2		
1.4 Coarse fritted impinger (no CFP)	Impinger	38.4 (94.1)	5.2	36.0	5.4
	Air blank from impinger	2.4 (5.9)	1.4		

assign nominal values to analyte present at low levels. In cases where a sample result was below the limit of detection (LOD), the average of the value zero (0) and the LOD was used in the sample statistic calculation. In cases where a sample result was between the LOD and the limit of quantitation (LOQ), the average of the LOD and the LOQ was used in the sample statistic calculation.

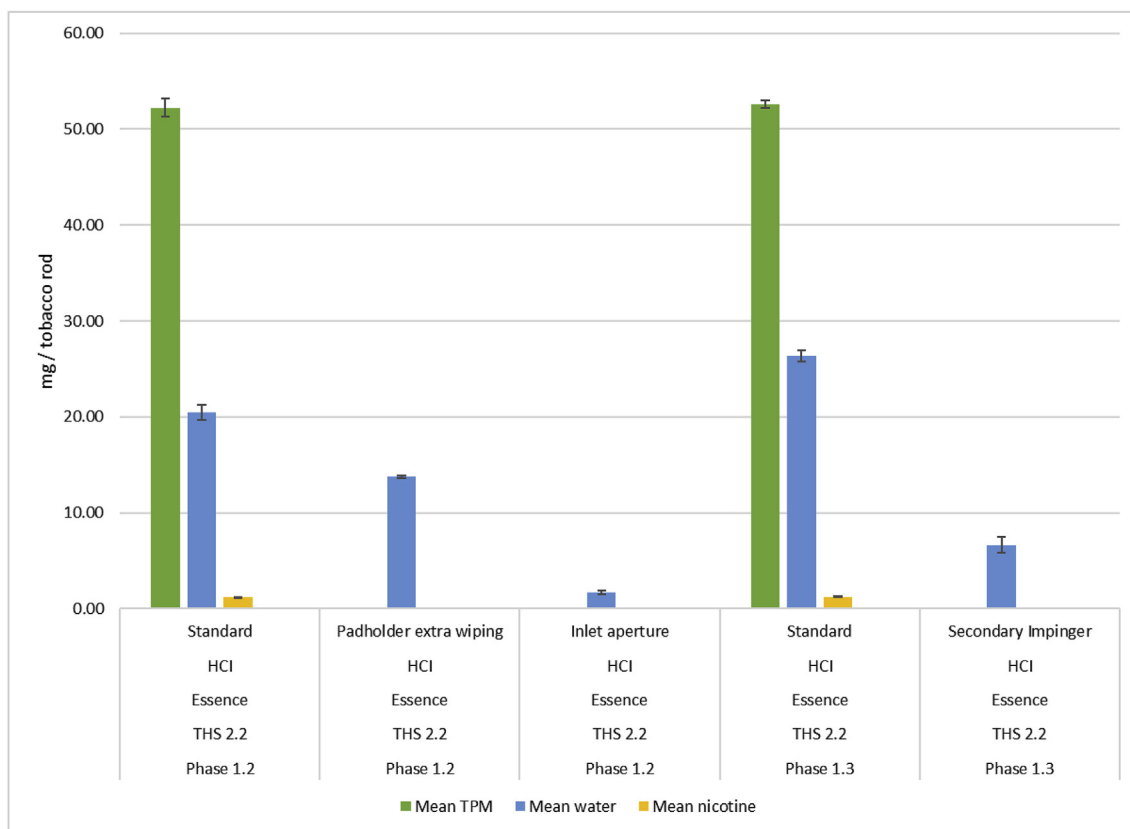
### 3. Results and discussion

The study was conducted in three phases (labelled numerically under Test column, Table 1), with phase 1 and 2 being exploratory to ascertain relevant parameters, and phase 3 consisting of further replication and validation using the set of most suitable parameters identified in phases 1 and 2 and confirming their efficacy. The work focused on the results obtained for TPM, water and nicotine, and the consequent correction to give NFDPM.

#### 3.1. Study phase 1

The results for the first phase are presented in Table 3, and graphically in Figs. 2 and 3 using THS 2.2 only. Additional wiping of the front and back parts of the pad holders, including the inlet aperture region (method 1.2) assessed whether total water recovery could be improved compared to the original method (1.1). The level of water vapour passing through the CFP was assessed using an impinger in series behind the CFP assembly (method 1.3). This was compared to a variant where the aerosol was captured only by the impinger (method 1.4). The values recorded for the impingers are after correction for the water levels present in the respective air blanks.

The individual component results in Fig. 2 show that water collected on the pad holder in areas that would not be swabbed under standard methodology. Additional wiping, specifically of the back of the pad holder led to a large increase in water recovery. The deposits from the inlet aperture were proportionally less, but still large enough to be important. Nicotine deposits in these zones were small enough that their impact on gravimetric total mass was unimportant. There was also



**Fig. 2.** Individual component yields in THS 2.2 mainstream emissions during Phase 1 (Mean ± SE, n = 5) with differentiation by method, puffing regime, product type and study phase.

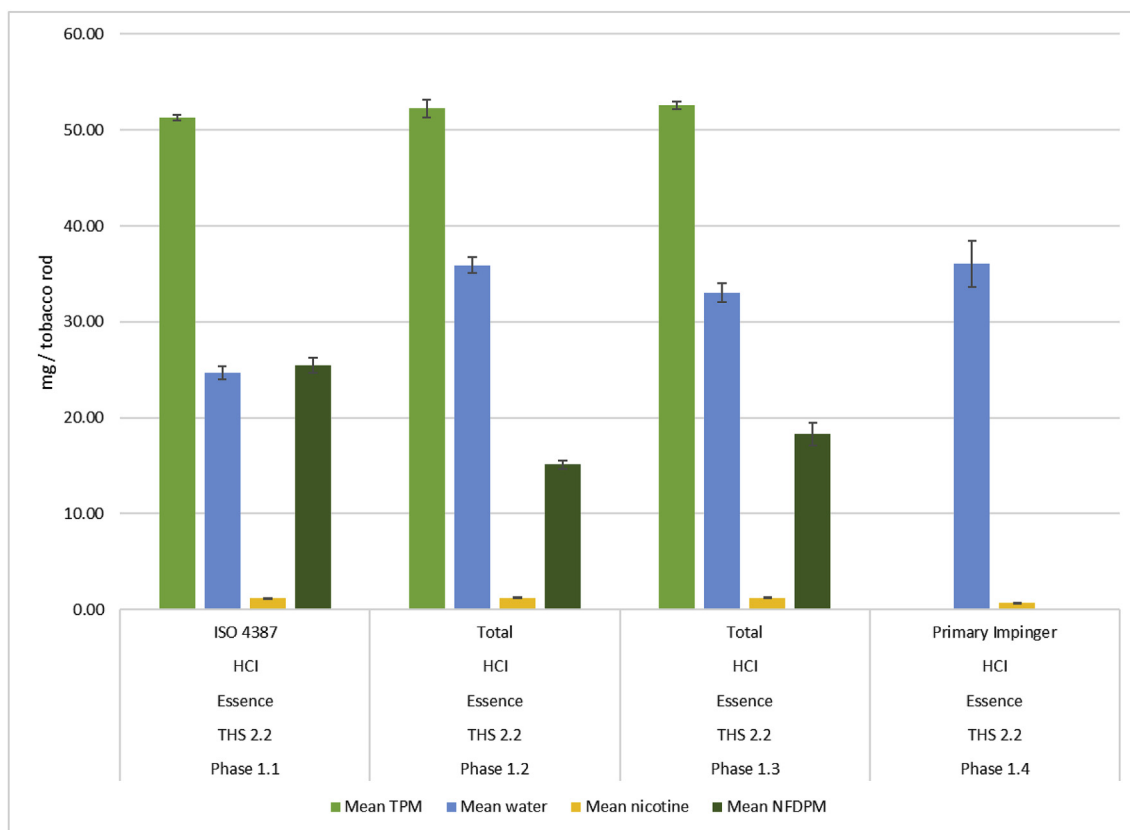


Fig. 3. Total yields in THS 2.2 mainstream emissions during Phase 1 (Mean ± SE, n = 5) with differentiation by method and study phase.

a small amount of water that passed through the CFP assembly and was trapped by the secondary impinger. This is the gaseous water that passed through the filter assembly and would not be captured by the gravimetric method.

The results for the total yields in Fig. 3 showed a large drop in calculated NFDPM values when the TPM value was adjusted for further water levels present in the filter assembly. The total composite water measurement was less than, but approaching, the figure determined by using an impinger for primary capture. Taken together this gives reasonable assurance that the sampling procedure can be adequately improved for the condensation and capture of a large proportion of HTP water by adapting procedures targeting areas that have not routinely been specified in the original standard.

### 3.2. Study phase 2

The results for the second phase study are presented in Table 4 and Fig. 4. This phase explored further possible sources of water losses, for example, changes that could be made to minimise evaporative water losses during sample generation and recovery. The CFP assembly (Fig. 1) has close-fitting covers (front and back caps, only one is shown

for illustration purpose) with o-rings that are designed to prevent evaporative losses or hygroscopic ingress (the latter, for example, as a result of a high level of glycerol present in HTP aerosol). Instructions were given to perform the standard method with extra care to apply these covers immediately after puffing, and to minimise any time periods where water could be transferred in or out of the system during CFP recovery and extraction. Also, liquid droplets have been noted on these covers when testing HTP products. They form part of the weighed mass for the filter and are not routinely wiped, therefore new procedures were used to swab them at this stage. The inlet labyrinth seals are in a separate carrier-housing which is detached prior to weighing the filter unit, however they are noted to sometimes become wet during HTP testing. Hence these were also included in the experiment.

The presence of water and traces of nicotine in most of these additional sampling confirmed the findings from the phase 1 study. Condensate was present on the pad holder and inlet aperture. Low levels of analyte were also sporadically present on the plastic covers. Levels in the labyrinth seals are worth noting as another possible limitation to the standard methodology. These observations may also have wider implications for other condensed phase analytes.

Table 4

Mainstream water yields in THS 2.2 emissions during Phase 2 (n = 5). Values in bracket are percentage of water in each component.

Method	Part analysed	Mean (mg/tobacco rod)	SD	Total water (mg/tobacco rod)	SD
2.1 Standard method	standard method	29.8 (100)	4.0	29.8	4.0
2.2 Modified by extra care with capping plus additional wiping of front and back of holder, inlet aperture and labyrinth seals	Standard	24.0 (68.4)	2.1	35.1	2.3
	Holder and inlet aperture	8.2 (23.4)	2.1		
	Seals	1.8 (5.1)	1.1		
	Cover	1.1 (3.1)	0.8		

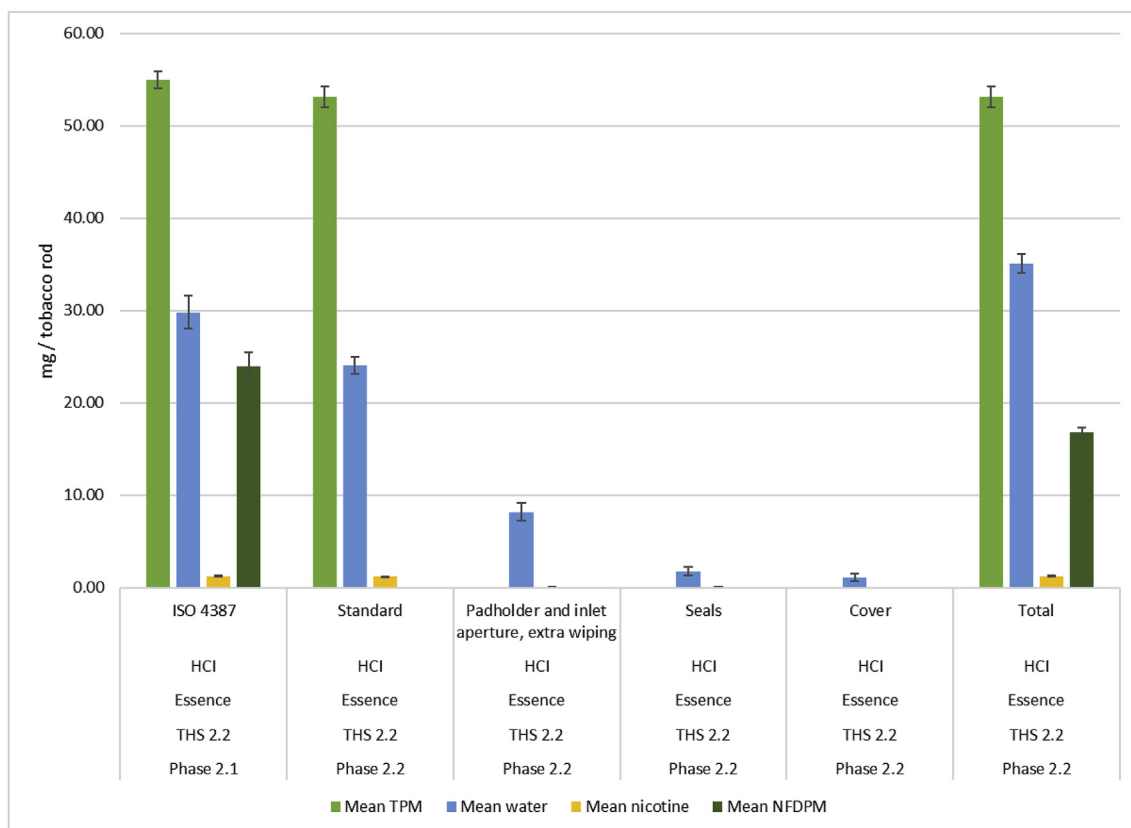


Fig. 4. Total and component yields for THS 2.2 mainstream emissions during Phase 2 (Mean ± SE, n = 5) with differentiation by method and study phase.

### 3.3. Study phase 3

The third phase of the study focussed on replication of the most relevant procedural modifications, and testing of some further product variants, including limited runs using ISO puffing for additional insight. The results are presented in Table 5 for THS 2.2 and Table 6 for THP 1.0, with Figs. 5 and 6 showing the breakdown of individual component yields for THS 2.2 and THP1.0 in Phase 3.3 and Fig. 7 showing the major trends in total recovery across all the product groups tested.

The data from phase 3.3 for individual components (Fig. 5) showed similar trends to those seen in phase 2.2. The total water yield for THS 2.2 was higher when summed over individual components in phase 3.3 than in the combined extraction in phase 3.2 (Fig. 6), which we attributed to the accumulation of errors. These data were intended for information purposes to show where water was deposited. The primary comparison for total levels was between Phase 3.1 and 3.2, which represented the standard and modified procedures. These data sets showed consistent TPM yields between the standard (Phase 3.1) and modified (Phase 3.2) (two sample *t*-test, *p* = 0.89, estimate for difference = -0.07 mg, 95% CI = -1.14 mg–1.00 mg). The difference between water levels using the standard and modified methods was highly

significant (two sample *t*-test, *p* < 0.000, estimate for difference = 6.74 mg, 95% CI = 5.28 mg–8.20 mg). This was reflected in the NFDPM (two sample *t*-test, *p* < 0.000, estimate for difference = -6.80 mg, 95% CI = -5.20 mg to -8.40 mg).

The water data for THP1.0 from the standard method were somewhat higher than previously published data (e.g. Forster et al., 2018). For THP1.0(T), the use of method 3.2 resulted in a small, but statistically significant increase in the measured water yield over the value from the standard method (two-sample *t*-test, *p* = 0.02, estimate for difference = 0.85 mg, 95% CI = 0.12 mg–1.58 mg). The increase was not significant for THP 1.0(M) (two-sample *t*-test, *p* = 0.82, estimate for difference = 0.12 mg, 95% CI = -0.97 mg–1.22 mg). In general, this highlights the substantially lower water yield of THP1.0 compared to THS 2.2, although both are still much higher than those reported for cigarette smoke. THP1.0 yields under ISO puffing parameters were included as a comparator for these main aerosol constituents using the standard ISO method, as for many regulatory laboratories HTPs are new product category and there is a lack of reported values for reference. Understandably the aerosol yield was very much lower than those obtained under the HCI puffing regime, reflecting the lower intensity of the puffing regime, and the smaller number of puffs overall. Note that

Table 5  
Mainstream water yields in THS 2.2 emissions during Phase 3 (n = 17). Values in bracket are percentage of water in each component.

Method	Part analysed	Mean (mg/tobacco rod)	SD	Total (mg/tobacco rod, by method)	SD
3.1 Standard	Standard	27.9 (100)	2.5	27.9	2.5
3.2 Modified = Standard plus additional wiping (Pad holder + covers and inlet aperture)	Combined extract	34.7 (100)	1.5	34.7	1.5
3.3 Same as 3.2, but separate extraction	Standard	28.8 (73.7)	1.6	39.1	1.4
	Holder	7.8 (20.0)	1.3		
	Inlet aperture	2.4 (6.1)	0.6		
	Cap	0.06 (0.2)	0.1		

**Table 6**  
Mainstream water yields in THP 1.0 emissions during phase 3, n = 17 (HCI puffing) or 5 (ISO puffing).

Method	Puffing/reps	Mean Bright Tobacco (mg/tobacco rod)	SD	Mean Intensely Fresh (mg/tobacco rod)	SD
3.1 Standard	HCI (n = 17)	13.4	0.9	13.5	1.2
3.2 Modified = Standard plus additional wiping (Pad holder + covers and inlet aperture)	HCI (n = 17)	14.3	1.2	13.6	1.9
3.4 Standard	ISO (n = 5)	4.9	0.4	5.8	0.9

there was a comparatively large difference in TPM value between the standard and modified test sets for THP1.0 which was highly significant in both variants; (for THP1.0(T) two-sample *t*-test,  $p < 0.000$ , estimate for difference = 3.21 mg, 95% CI = 2.17 mg–4.25 mg) and (THP1.0(M) two-sample *t*-test,  $p < 0.000$ , estimate for difference = 4.11 mg, 95% CI = 2.66 mg–5.57 mg). This could be due to within-batch product variability plus any method weakness.

3.4. Impact on NFDPM calculations

Mainstream aerosol yields for THS 2.2 and THP1.0(T) and THP1.0(M) are shown in Table 7. For THS2.2, TPM and nicotine yields were consistent across experiments, and the increased water yield measured using the modified method is reflected in the reduced NFDPM. For THP1.0, the TPM yields were less consistent than the nicotine yields, and the increases in water yield arising from the modified method seemed less consequential. Limited data for THP1.0 yields under the ISO puffing parameters was obtained as a comparator but the water yield was not determined using the modified method.

A comparison can be made between the product types and variants if we normalise NFDPM as a fraction of TPM. Fig. 8 shows NFDPM/TPM ratio values across the two HTP products and plus literature reported

results from 3R4F reference cigarette. They show a relatively consistent picture for HTPs, with data stratified according to the different pad treatment methods, rather than the different puffing regime or the product type. The HTP results are clearly different to those reported for the 3R4F, consistent with the markedly different physiochemical nature of the aerosol between the two product categories.

In summary, we propose modification to perform standard extraction and wiping of the CFP assembly, with additional wiping as follows: wipe the inner surface of the holder using additional  $2 \times 1/4$  pads, wipe the inner surface of the front aperture of the pad holder (termed the inlet aperture, which is the normal position of the neoprene washer that is not used) using a further  $1/4$  pad, plus wipe the inner surface of the front and back aperture covers for the pad holder using another  $1/4$  pad. The main pad and all 6 quarter pad segments are to be extracted together, with suitable correction to be made to the background standardisation and calibration procedures, to compensate for the additional pads. From the point of setting a standard method, these extra steps will make the ISO 4387 Standard into a new standard, purposed for HTPs. We do not propose for these extra steps to be included in the ISO 4387 Standard to test cigarette smoke, as the Standard has been proved to be adequate for this purpose.

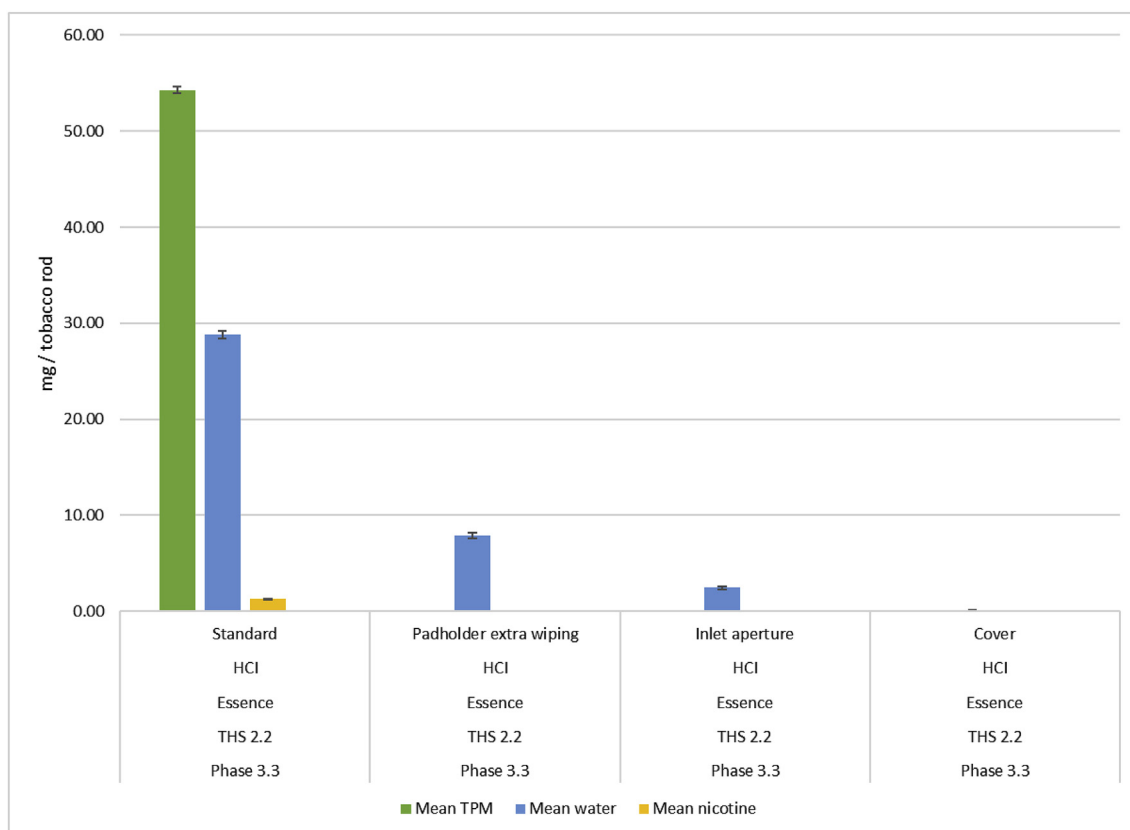


Fig. 5. Component yields for THS 2.2 mainstream emissions during Phase 3.3 (Mean ± SE, n = 17) with differentiation by method and study phase.



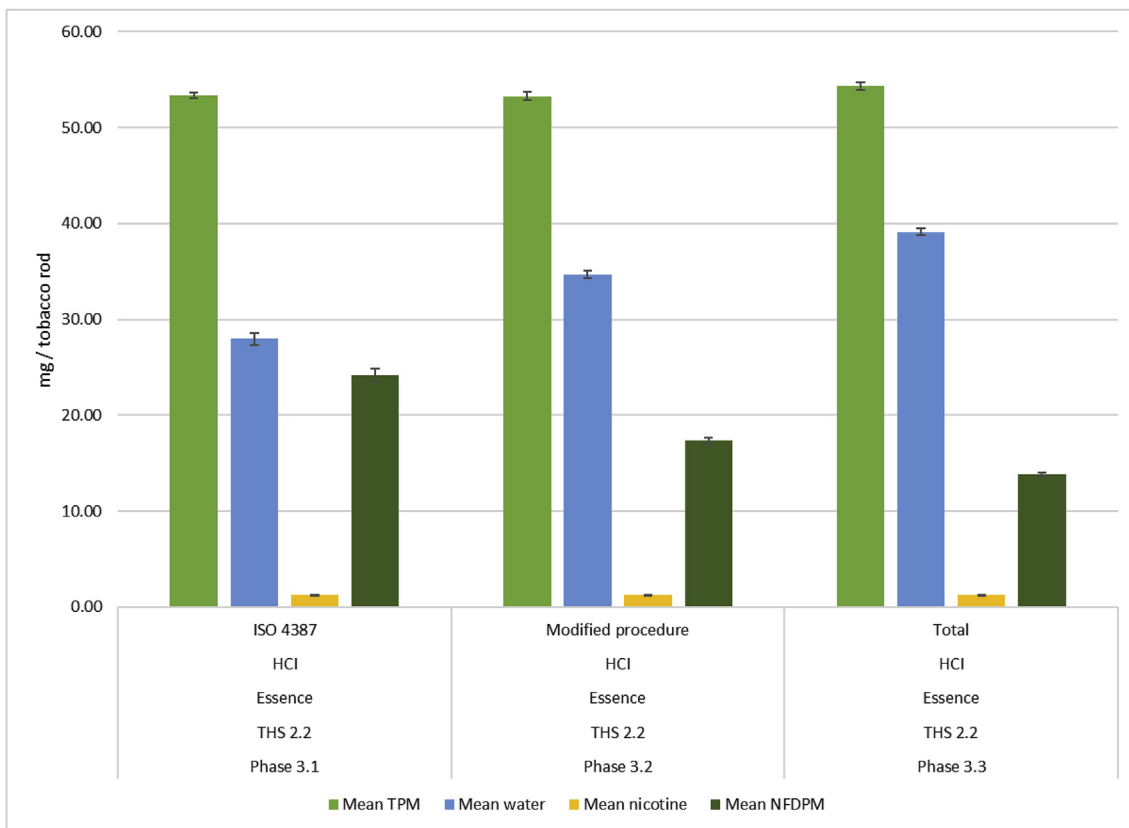


Fig. 6. Total yields for THS 2.2 mainstream emissions during Phase 3 (Mean ± SE, n = 17) with differentiation by method and study phase.

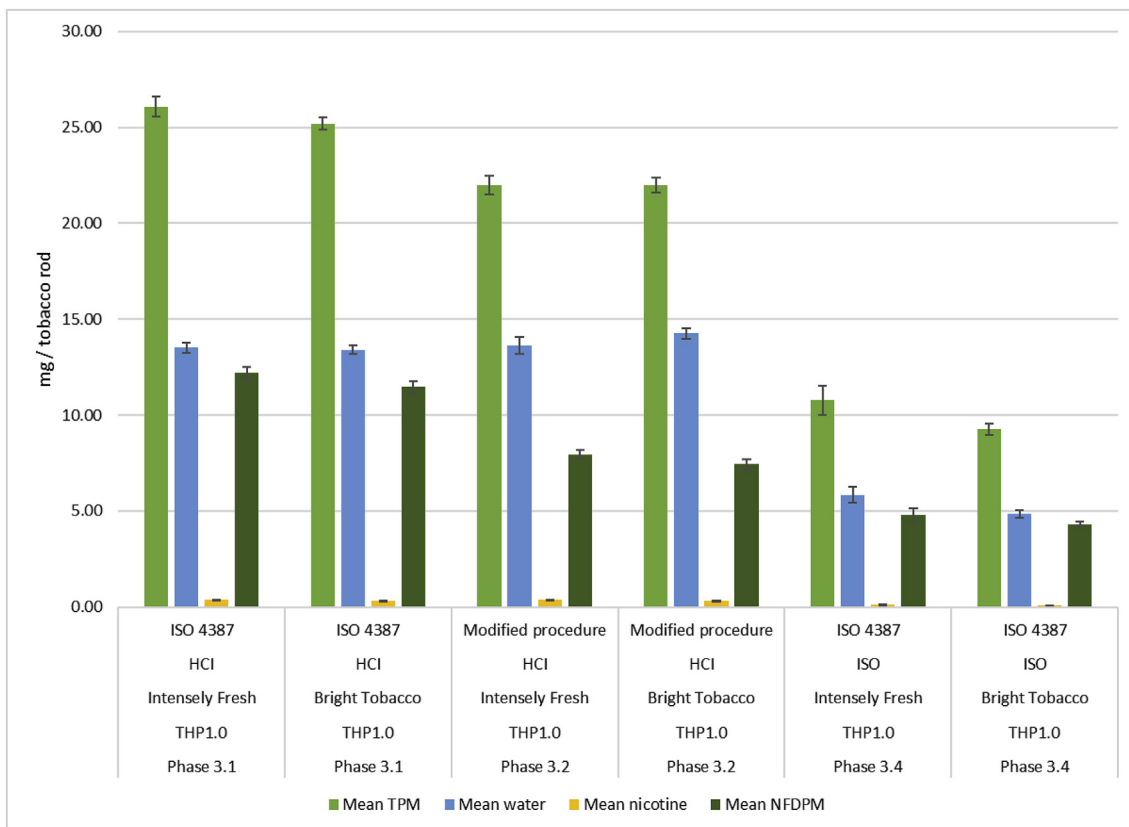


Fig. 7. Total yields for THP 1.0 mainstream emissions during Phase 3 (Mean ± SE, n = 17 under HCl puffing and n = 5 under ISO puffing) with differentiation by method, puffing regime, product type and study phase.

**Table 7**  
Mainstream yields for THS 2.2 and THP 1.0 under standard and modified methods.

Product	Puffing	Method	Mainstream yields mg/tobacco rod, Mean (SD)				
			TPM	Nicotine	Water	NFDPM	NFDPM/TPM (%)
THS 2.2	HCI	1.1 Standard	51.3 (0.6)	1.14 (0.05)	24.7 (1.6)	25.5 (1.7)	49.7
		2.1 Standard	55.0 (2.1)	1.24 (0.10)	29.8 (4.0)	24.0 (3.3)	43.6
		3.1 Standard	53.3 (1.2)	1.23 (0.05)	27.9 (2.5)	24.1 (2.8)	45.2
		3.2 Modified	53.3 (1.8)	1.22 (0.07)	34.7 (1.5)	17.3 (1.5)	32.5
		3.3 Modified	54.3 (1.6)	1.29 (0.05)	39.1 (1.4)	13.9 (0.8)	25.6
THP1.0(T)	HCI	3.1 Standard	25.2 (1.4)	0.31 (0.03)	13.4 (0.9)	11.5 (1.2)	45.6
		3.2 Modified	22.0 (1.6)	0.31 (0.04)	14.3 (1.2)	7.4 (1.0)	33.6
		3.4 Standard	9.3 (0.7)	0.09 (0.01)	4.9 (0.4)	4.3 (0.3)	46.2
THP1.0(M)	HCI	3.1 Standard	26.1 (2.1)	0.36 (0.03)	13.5 (1.2)	12.2 (1.3)	46.7
		3.2 Modified	22.0 (2.1)	0.37 (0.04)	13.6 (1.9)	8.0 (0.8)	36.4
		3.4 Standard	10.8 (1.7)	0.13 (0.02)	5.8 (0.9)	4.8 (0.8)	44.4

3.5. Implications for HTP aerosol

The results obtained from this work need to be examined to compare the thermophysics behind the smoke formation in cigarette and aerosol generation from HTPs (Fig. 9). For cigarette smoke, this is well studied and can be best illustrated by a schematic (Baker, 1999). Briefly, cigarette combustion is a process where tobacco burns in the presence of air and an initiating ignition source. The burning process is an exothermic, self-sustaining reaction that consumes the solid tobacco and produces a highly complex mixture of gases and smoke particles, leaving behind a residual inorganic ash. The different mechanistic steps interact and modulate due to the presence or absence of an external puff, alternating between the state of smouldering and puffing burn. Smouldering burn occurs around 650 °C and consumes a larger portion of the tobacco rod, emitting the sidestream smoke. Puffing burn causes the temperature of the burning coal to rise rapidly (with maximum temperature beyond 900 °C) and produces mainstream smoke. The relative concentrations of main classes of chemicals produced from smouldering and puffing under standard machine smoking conditions are different (Baker, 1974).

Electrically heated tobacco heating products are designed to work differently (Fig. 9) (Eaton et al., 2018; Schaller et al., 2016a). The main sensorial aerosol comes from added aerosol agent such as glycerol (Forster et al., 2018; Schaller et al., 2016b), which can be up to 15% of the tobacco material used. The predominate release of water to the

mainstream during puffing, exacerbated by glycerol's affinity to attract moisture, contribute to a high reported water content in the HTP aerosol, irrespective of any modification to the aerosol trapping procedures. The maximum heating temperature setting in the currently available commercial HTPs is around 350 °C and the speed of energy supply via a resistive heating mechanism prevents any active energy compensation during a 2-s puff. Therefore the tobacco in HTPs undergoes a significant temperature drop (~50 °C or more) when a puff is taken and the ambient air cools the tobacco, along with temperature drop due to evaporation of volatiles (Eaton et al., 2018). This is in clear contrast to a greater than 200 °C rise in a burning cigarette (Liu and Woodcock, 2014). The result is that tobacco combustion in HTPs is eliminated and tobacco pyrolysis is reduced to a minimum (Eaton et al., 2018; Schaller et al., 2016b). The dominating aerosol formation routes in HTPs are evaporation and distillation. The main aerosol constituents (water and glycerol) in the tobacco leaf will preferentially utilise the thermal energy, followed by an initial thermal distillation of the tobacco. Hence, we propose that this aerosol entity distilled out of the tobacco, which may be of product performance and regulatory interests, be called “distillate”. A larger portion of evaporative water is present in the HTP aerosol stream than that found in cigarette smoke, and its dynamic equilibrium during puffing and aerosol capture, present a challenge to the ISO 4387 Method, as demonstrated by the results obtained in this work.

The composition of the particulate phase of HTP1.0 is shown in

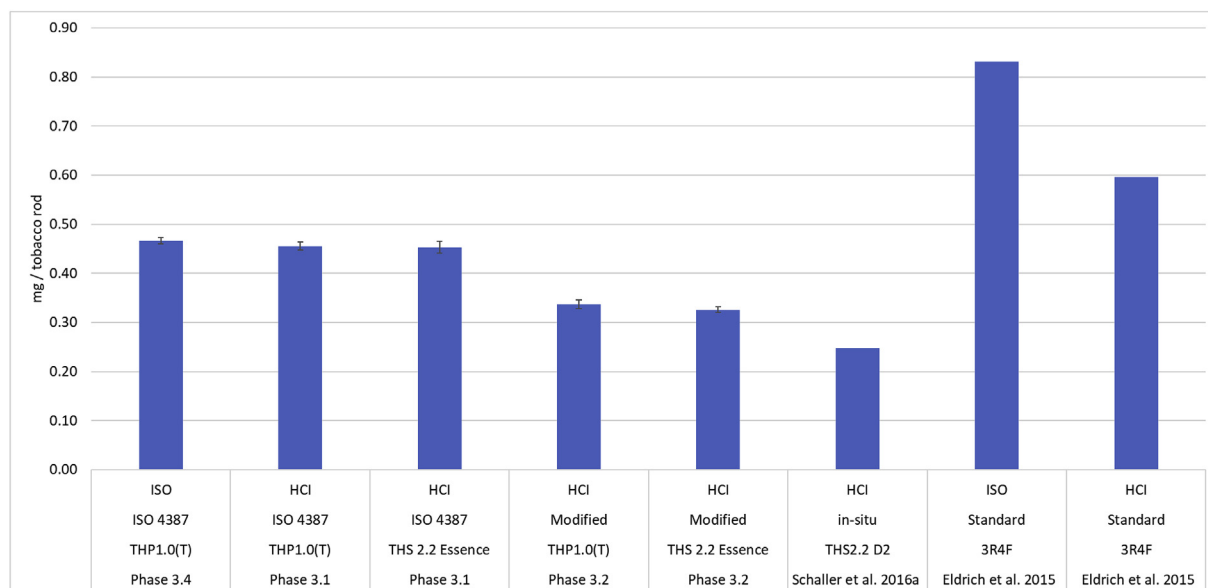


Fig. 8. NFDPM/TPM ratios allowing normalisation and comparison across product categories.

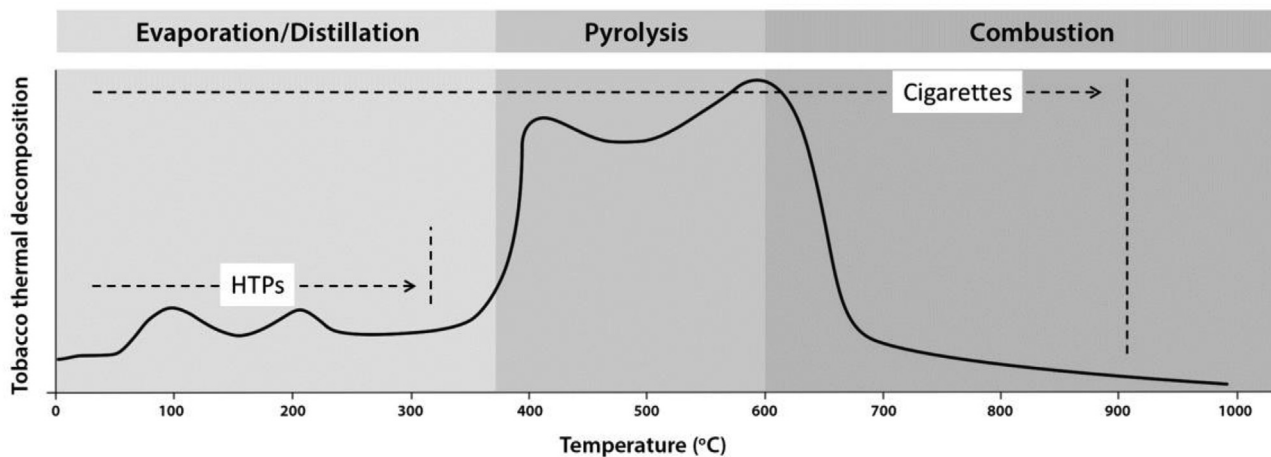
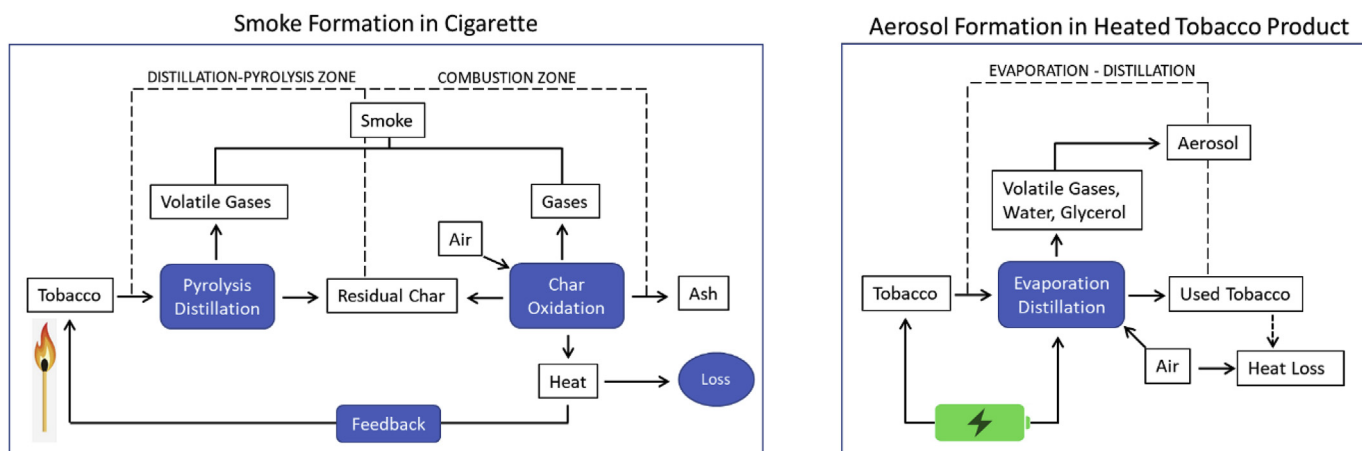
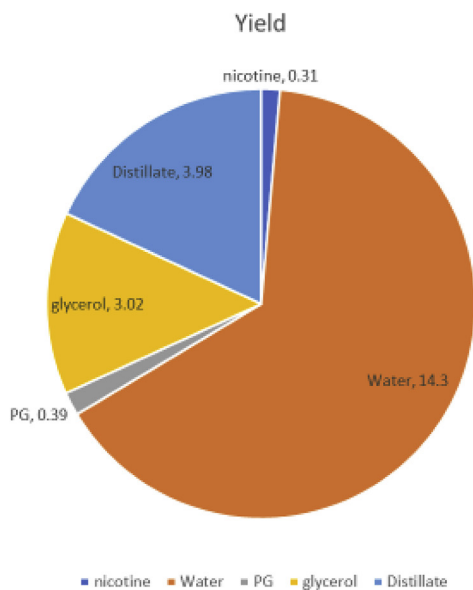


Fig. 9. Thermophysical difference between cigarette smoke (Baker, 1999) and HTP aerosol formation. Peaks around 100 and 200 °C used to illustrate water and glycerol release, and are variable depending on the material and heating rate.



**Mainstream constituents**

Parameter	Yield (mg/Neostik)
TPM	22.0
Nic	0.31
Water	14.3
NFDPM	7.4

**NFDPM beakdown**

Parameter	Yield (mg/Neostik)
NFDPM	7.4
Glycerol	3.02
PG	0.39
Distillate	3.98

Fig. 10. A breakdown of main aerosol composition from a THP1.0.

Fig. 10 (obtained by the modified procedure and under HCI puffing regime). The majority of the mass is water. The level of unspecified distillate is relatively small, at around 4 mg per tobacco rod. Taken together, the results shown in this work and the body of knowledge on cigarette smoke analyses, illustrate that the terms defined and used to describe cigarette smoke aerosol, such as TPM and NFDPM, cannot be extrapolated into different product categories, such as HTPs. Given the fundamental differences in the composition and properties of the aerosol entities between cigarette smoke and HTP aerosol, comparisons between values of these parameters could be highly misleading.

#### 4. Conclusions

A 3-phase study has investigated key parameters and their modifications to the standard ISO method for NFDPM determination in mainstream HTP emissions, especially around the aerosol extraction. The results have demonstrated that procedural modifications can be made to the standard ISO 4387 method to a practically usable extent to recover water deposits from areas that are not routinely specified. The results also highlighted chemical and physical differences in aerosol properties from HTPs, which are consistent with the understanding that these products work differently to cigarettes. Product testing and HTP-specific standard development should consider these fundamental differences and ascertain whether cigarette smoke derived methods are suitable for the novel product category. In addition, care should be taken to describe HTP aerosol properties using cigarette smoke derived terminology (e.g., tar), which does not reflect the aerosol formation principle and could lead to erroneous practices of adopting existing standard methods without method validation. By its chemical and physical nature, the NFDPM from HTP aerosol is more appropriately described as “distillate”. The modification in Phase 3.2 offers a suitable interim method to allow ISO 4387 to be used for this emerging product category by a competent laboratory. Further cross-laboratory tests are needed to establish the method's repeatability and reproducibility.

#### Conflicts of interest

The work reported in this publication was solely funded by British American Tobacco Investments Ltd. All authors are employees of BAT except for JN who worked under contractual agreements.

#### Transparency document

Transparency document related to this article can be found online at [doi:10.1016/j.yrtph.2018.09.016](https://doi.org/10.1016/j.yrtph.2018.09.016).

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