Beiträge zur Tabakforschung International/*Contributions to Tobacco Research* Volume 24 · No. 3 · November 2010

DOI: 10.2478/cttr-2013-0891

Short Communication

Influence of Tobacco Additives on the Chemical Composition of Mainstream Smoke - Additional Analysis of Three Tobacco Industry Based Laboratories*

by

Michael Intorp [1], Jutta Pani [2], Martin Blumenstock [3]

[1] Imperial Tobacco Group, Albert-Einstein-Ring 7, 22761 Hamburg, Germany
[2] JTI/Ökolab GmbH., Hasnerstrasse 127, 1160-Vienna, Austria
[3] British American Tobacco, Product Centre Europe, Weiherstraße 26, 95448 Bayreuth, Germany

SUMMARY

Three tobacco industry based laboratories determined selected mainstream components using their established in-house methods. Machine smoking was done according to the ISO smoking regime. The Test cigarettes smoked for this investigation were manufactured with different amounts of added glycerol, cocoa powder and sucrose.
Variability between the three laboratories differed clearly for the analyzed smoke components. No overall effects due to the added ingredients on smoke components could be found. The high 'tar' products with the highest lodading of sucrose showed a slight increase in formaldehyde emissions among all three laboratories. [Beitr. Tabakforsch. Int. 24 (2010) 139–144]

ZUSAMMENFASSUNG

Drei Laboratorien der Zigarettenindustrie bestimmten ausgewählte Hauptstomrauchbestandteile mit Hilfe ihrer in den Häusern etablierten Methoden. Das maschinelle Abrauchen erfolgte entsprechend den ISO-Bedingungen. Die für diese Untersuchung eingesetzten Zigaretten waren mit unterschiedlichem Zusatz von Glycerin, Kakaopulver und Saccharose hergestellt worden.
Es wurden deutliche Unterschiede zwischen den beteiligten Laboratorien hinsichtlich der analysierten Rauchkomponenten festgestellt. Es konnten keine allgemeinen Auswirkungen der zugesetzten Stoffe auf die Zusammensetzung des Hauptstomrauches nachgewiesen werden. Zigaretten mit einem höheren 'Teer'-Gehalt und der höchsten Zugabe von Saccharose zeigten einen leichten Anstieg der Formaldehyd-Konzentrationen in allen drei Laboratorien. [Beitr. Tabakforsch. Int. 24 (2010) 139–144]

RESUME

Trois laboratoires industriels ont quantifié des composés de la fumée principale avec leurs propres méthodes d'analyses en utilisant le régime de fumage ISO. Les cigarettes testées ont été fabriquées avec différentes quantités de glycérine, de poudre de cacao et de saccharose. Pour les composés de la fumée analysés, la variabilité entre les trois laboratoires est clairement différente. L'ajout des ingrédients n'a montré aucun effet sur la concentration de ces composés. Seul l'ajout le plus élevé de saccharose sur les produits donnant des "Tars" importants montre, pour les tous les laboratoires, une légère augmentation dans la production de formaldéhyde. [Beitr. Tabakforsch. Int. 24 (2010) 139–144]

INTRODUCTION

The following communication contains the results of mainstream smoke analysis of the test cigarettes (test pieces) already described and characterized in the paper of

*Received: 10th March 2010 – accepted: 18th September 2010

Table 1. Methods routinely used by the three laboratories for measuring the analytes under consideration.

Parameter	Lab 1	Lab 2	Lab 3
1,3-Butadiene	Health Canada T-116 (gas phase sampling in cooled impingers followed by GC-MS analysis) (5) [a]	In-house method; gas bag sampling followed by GC-FID analysis	Health Canada T-116, but GC-Ion trap
Isoprene	Health Canada T-116 (gas phase sampling in cooled impingers followed by GC-MS analysis)	In-house method; gas bag sampling followed by GC-FID analysis	Health Canada T-116 but GC-Ion trap
Formaldehyde	Health Canada T-104 (HPLC-UV detection) (5)	In-house method based on Health Canada T-104	Health Canada T-104 (HPLC-UV detection)
Acetaldehyde	Health Canada T-104 (HPLC-UV detection)	In-house method based on Health Canada T-104	Health Canada T-104 (HPLC-UV detection)
TSNA (NNN, NNK, NAT, NAB)	Health Canada T-111 (GC-TEA) (5)	In-house LC-MS/MS method	CORESTA Recommended Method No. 63; GC-TEA (6)
Benzo[*a*]pyrene	CORESTA Recommended Method No. 58; GC-MS method (7)	In-house method based on Health Canada T-103 (HPLC fluorescence detection) (5)	CORESTA Recommended Method No. 58; GC-MS method

[a] GC = Gas chromatography, MS = Mass spectrometry, FID = Flame ionization detector, HPLC = High-perfomance liquid chromatophraphy

HAHN and SCHAUB in this issue of *Beiträge zur Tabakforschung International* (1). The investigation was performed by the three laboratories BAT (Regulatory Analysis Laboratory, Souza Cruz), JTI (Ökolab Vienna), and Reemtsma (Hamburg).

For reasons of consistency the analytes measured in this investigation are the same as those selected at the beginning of the project in the study protocol of HAHN and SCHAUB. Machine smoking of the test pieces was carried out according to the ISO smoking regimen (2) as done by HAHN and SCHAUB.

ANALYTICAL METHODS

The laboratories, which participated in this study are all experienced in the analysis of 'Hoffmann analytes' and all of them are accredited according to ISO 17025 (3) for this group of smoke emissions. They also took part in the comprehensive collaborative study on the determination of 'Hoffmann analytes', organised by CORESTA´s Special Analytes Taskforce in 2006, which was recently published in *Beiträge zur Tabakforschung International* (4).

The three participating laboratories were asked to assess the emissions in the mainstream cigarette smoke of the test cigarettes with the methods routinely used for measuring these analytes. Table 1 gives an overview on the methods applied in each laboratory.

For evaluating results the repeatability and reproducibility of a method (8) are of major importance. This kind of data is available for the standardized methods for the determination of benzo[*a*]pyrene (B*a*P) and the tobacco-specific nitrosamines (TSNAs). The data are shown in Table 2.

RESULTS

An independent statistician collected the analytical data and processed the in a non-proprietary way. The results are shown in Figures 1–9 (see pages 142–144).

CONCLUSIONS

The results found for the individual analytes reflect the situation concerning the availability of International Standard Methods at that time. For the individual TSNA's smoke emission data obtained for these series of test cigarettes, similar levels were detected by all the laboratories for the individual products. The same tendency was observed for B*a*P measurement results. It should be noted that for TSNA and B*a*P, International Standard Methods are available as indicated in Table 1. These are CORESTA Recommended Methods (CRM) No. 63 for TSNA and CRM No. 58 for B*a*P. Increased levels of TSNAs or B*a*P in smoke were not observed that could have been caused by admixed amounts of the additives examined in the study.

In case of 1,3-butadiene the yields determined for the same products differ significantly among the laboratories. This can probably be attributed to the fact that different methods

Table 2: Repeatability and reproducibility data.

Parameter	Method	Cigarette	Mean (ng/cig)	Repeatability	Reproducibilty
B*a*P	CRM 58 (7)	Ky2R4F	7.28	1.27	2.52
NNK	CRM 63 (6)	Ky2R4F	141.39	15.68	43.88
NNN	CRM 63	Ky2R4F	146.01	10.75	32.26
NAT	CRM 63	Ky2R4F	143.38	20.38	63.62
NAB	CRM 63	Ky2R4F	16.60	5.15	10.70

in sampling and measuring had been used. However, even by only evaluating the results within the laboratories neither for the type of ingredient, nor for their applied levels, an increase of butadiene was observed.

The isoprene yields are found to be much more comparable among the laboratories and similar trends were seen in terms of ingredients and their amounts applied on the test cigarettes. But in this case too, no effect of added ingredients altering isoprene smoke yields was detected.

Further efforts in standardisation of a suitable method are urgently needed to obtain reliable reproducibility and repeatability data for both compounds, permitting to discriminate results obtained for different types of cigarettes sufficiently. In the meantime a CRM for selected volatiles was developed and published (9) however reproducibility and repeatability data are still on the level observed in this study.

The carbonyls formaldehyde and acetaldehyde are both determined by measuring their di-nitro-phenyl hydrazones with HPLC-UV or HPLC-DAD (diode array detection), the methodology being based on Health Canada Method T-104. But for both compounds considerable differences in yields were observed between laboratories. Due to the poor reproducibility a carbonyl method, too, would benefit greatly from an internationally acknowledged standardisation process.

It can be said, however, that all laboratories demonstrated a slight increase of formaldehyde for the 10 mg 'tar' cigarette with the highest load of sucrose. This observation was not confirmed for the 6 mg 'tar' product.

REFERENCES

1. Hahn, J. and J. Schaub: Influence of Tobacco Additives on the Chemical Composition of Mainstream Smoke. Beitr. Tabakforsch. Int. 24 (2010) 100–116.
2. International Organization for Standardization (ISO): ISO 3308:2000 Routine analytical cigarette-smoking machine - Definitions and standard conditions; ISO, Geneva, Switzerland, 2000.
3. International Organization for Standardization (ISO): ISO/IEC 17025:2005 General requirements for the competence of testing and calibration laboratories; ISO, Geneva, Switzerland, 2005.
4. Intorp, M., S. Purkis, M. Whittaker, and W. Wright: Determination of "Hoffmann Analytes" in Cigarette Mainstream Smoke. The Coresta 2006 Joint Experiment; Beitr. Tabakforsch. Int. 23 (2009) 161–202.
5. Health Canada Methods (Accessed November 2010) ; http://www.hc-sc.gc.ca/hc-ps/tobac-tabac/legislation/reg/indust/method/index-eng.php#main.
6. CORESTA, Recommended Method No 63: Determination of Tobacco Specific Nitrosamines in Cigarette Mainstream Smoke – GC-TEA Method; CORESTA, 2005.
7. CORESTA, Recommended Method No 58: Determination of Benzo[*a*]pyrene in Cigarette Mainstream Smoke - Gas Chromatography-Mass Spectrometry Method; CORESTA, 2004.
8. International Organization for Standardization (ISO): ISO 5725-2 Accuracy (trueness and precision) of measurement methods and results - Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method (ISO 5725-2:1994 including Technical Corrigendum 1:2002); ISO, Geneva, Switzerland, 2002.
9. CORESTA, Recommended Method No 70: Determination of Selected Volatile Organic Compounds in the Mainstream Smoke of Cigarettes - Gas Chromatography-Mass Spectrometry Method, 2010.

Corresponding author:

Michael Intorp
Imperial Tobacco Group
Albert-Einstein-Ring 7
22761 Hamburg
Germany
Michael.Intorp@Reemtsma.de

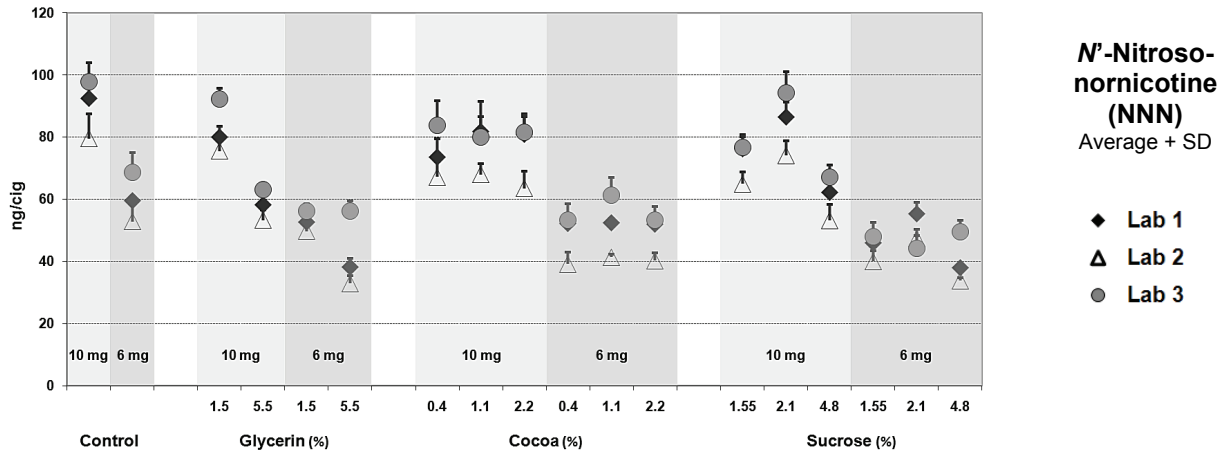

Figure 1. *N*'-Nitrosonornicotine (NNN) yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

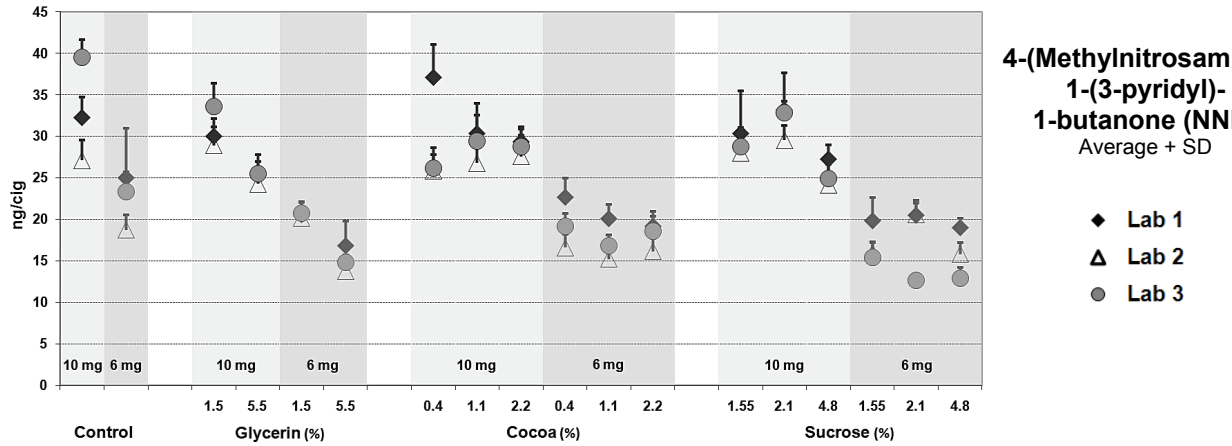

Figure 2. 4-(Methylnitrosamino)-1-(3-pyridyl)-1-butanone (NNK) yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

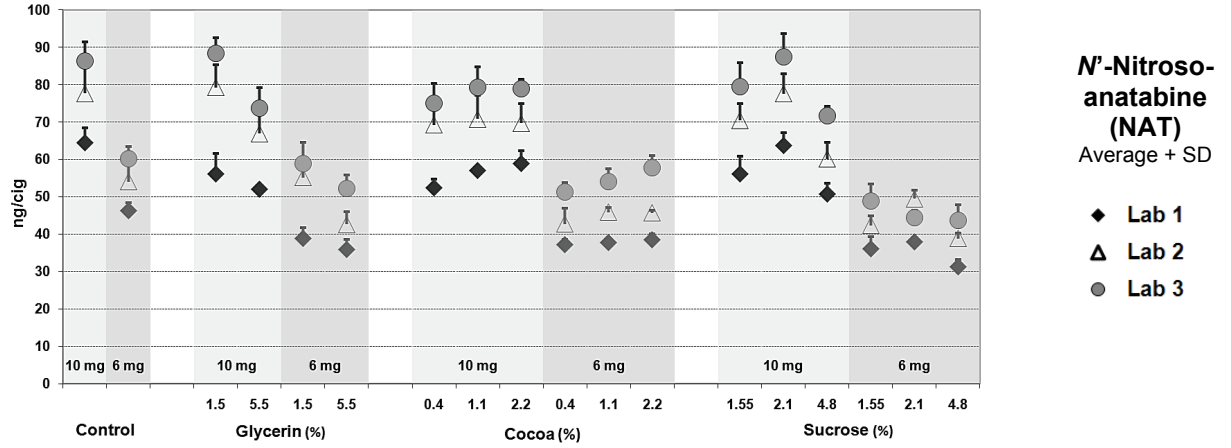

Figure 3. *N*'-Nitrosoanatabine (NAT) yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

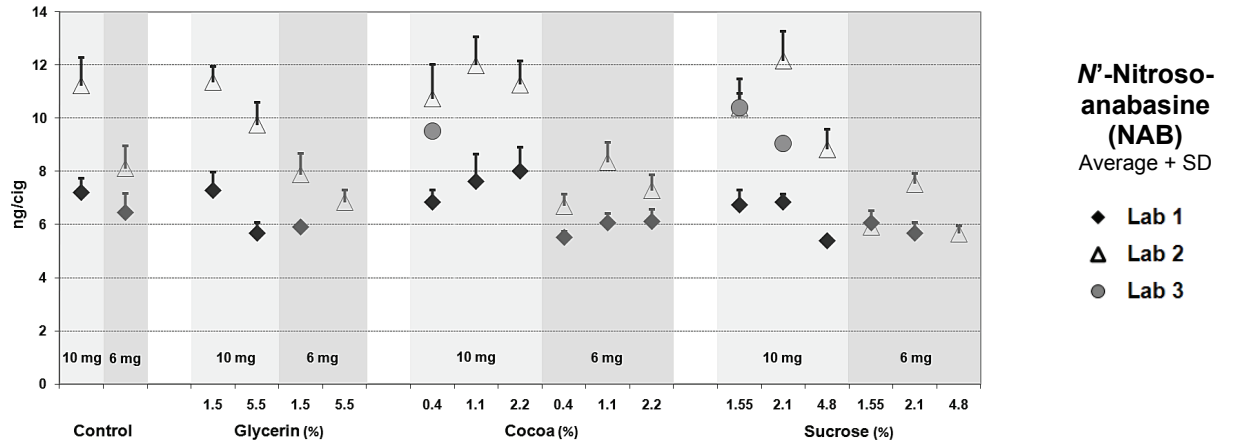

Figure 4. *N*'-Nitrosoanabasine (NAB) yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

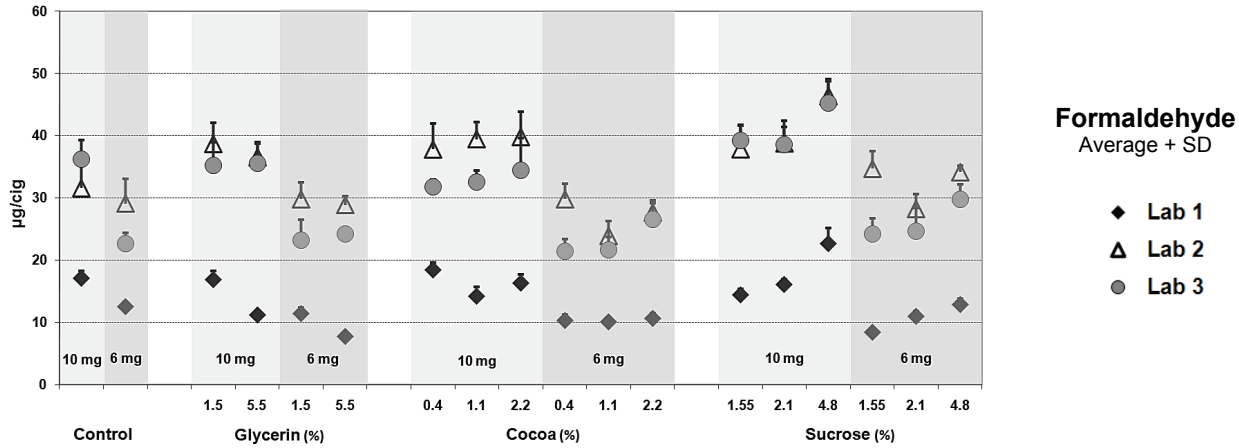

Figure 5. Formaldehyde yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

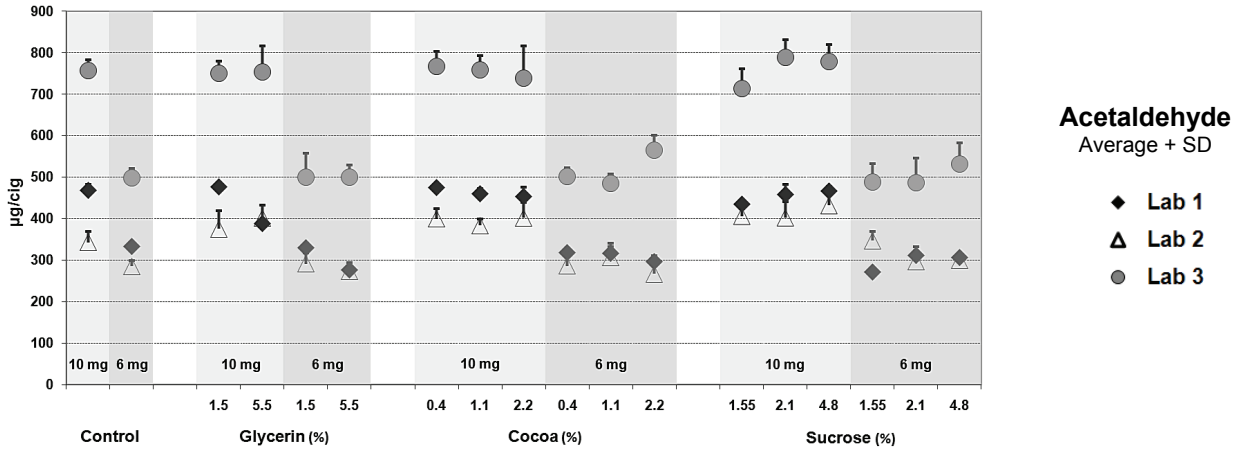

Figure 6. Acetaldehyde yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

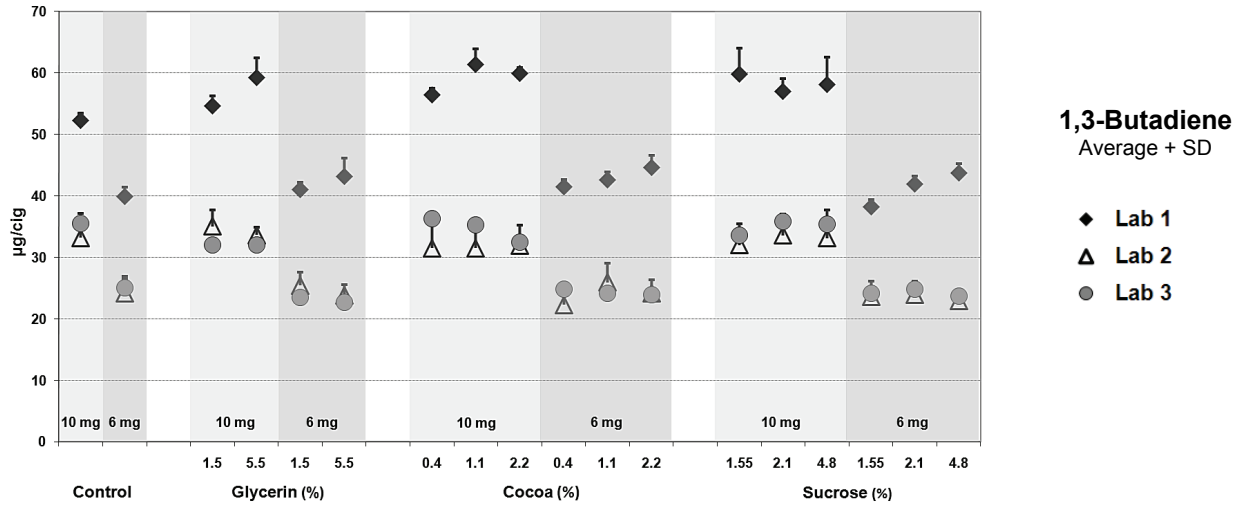

Figure 7. 1,3-Butadiene yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

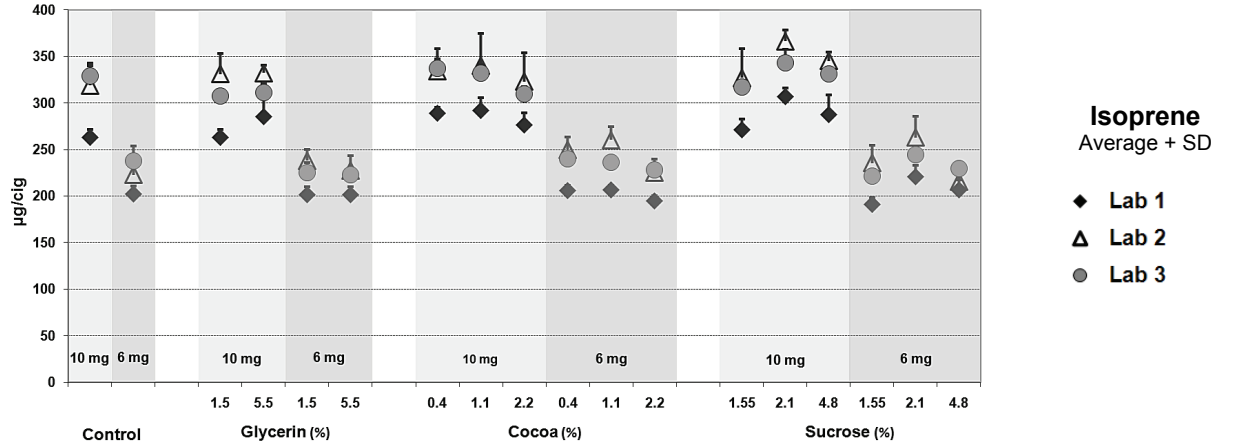

Figure 8. Isoprene yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.

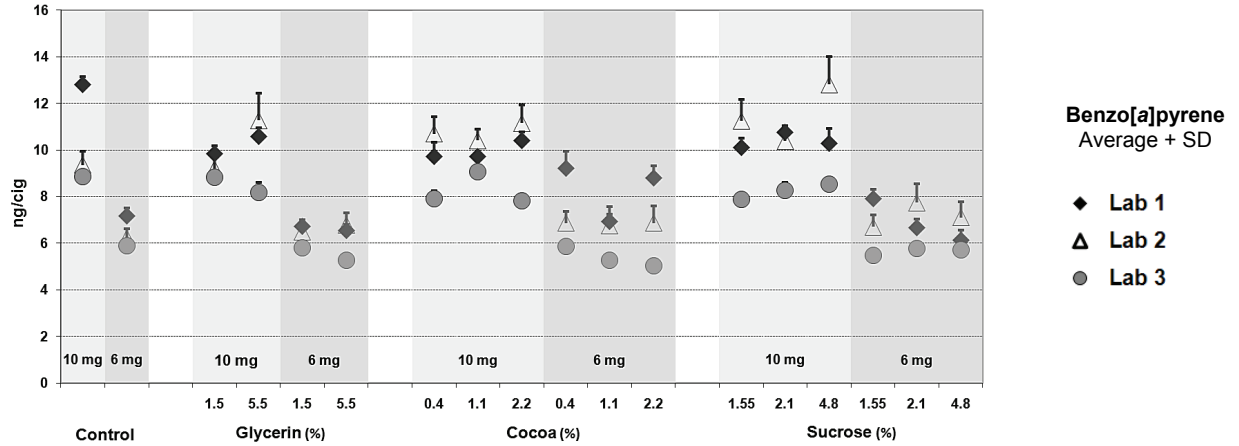

Figure 9. Benzo[*a*]pyrene yields in mainstream smoke for different levels of added glycerol, cocoa, and sucrose in two experimental cigarettes (10 mg, 6 mg nicotine-free dry particulate matter according to ISO smoking regime). Standard deviation (SD) is calculated for 5 replicates.