Modeling aerosol formation in an Electrically Heated Tobacco Product

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Outline

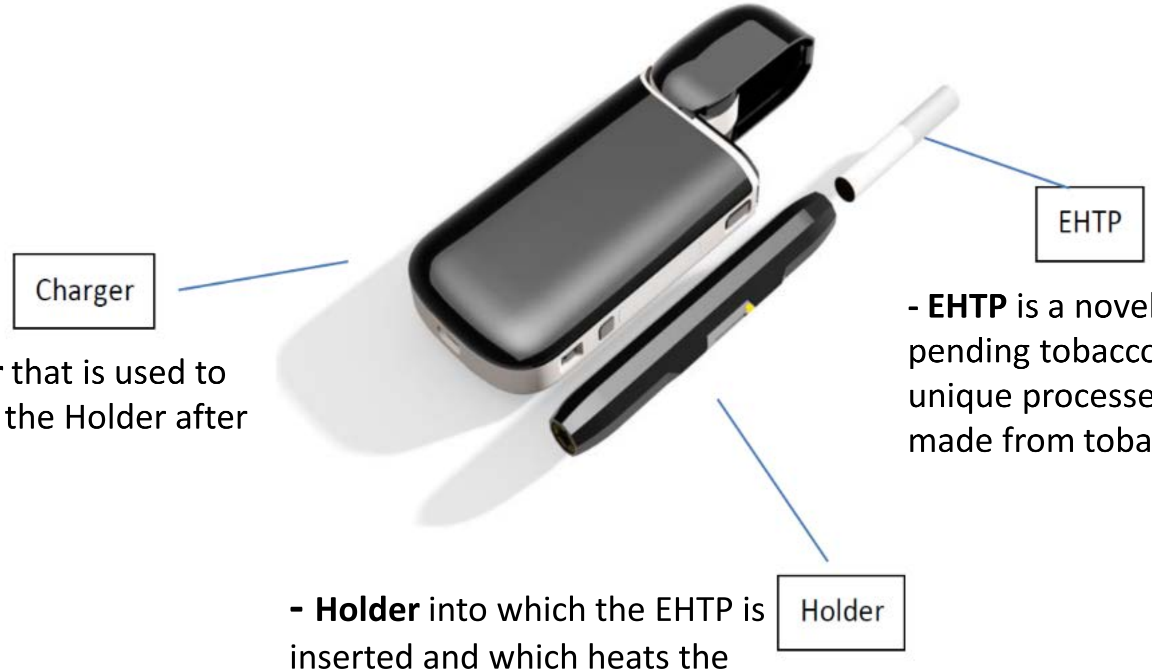
- Background & objective
- Modeling aerosol formation
- Simulation cases
- Results
- Conclusions

BACKGROUND
&
OBJECTIVE

What is an Electrically Heated Tobacco Product?

Electrically Heated Tobacco System (EHTS)
(also referred to as Tobacco Heating System (THS-2.2))

EHTS is composed of a charger, holder and electrically heated tobacco product (EHTP)



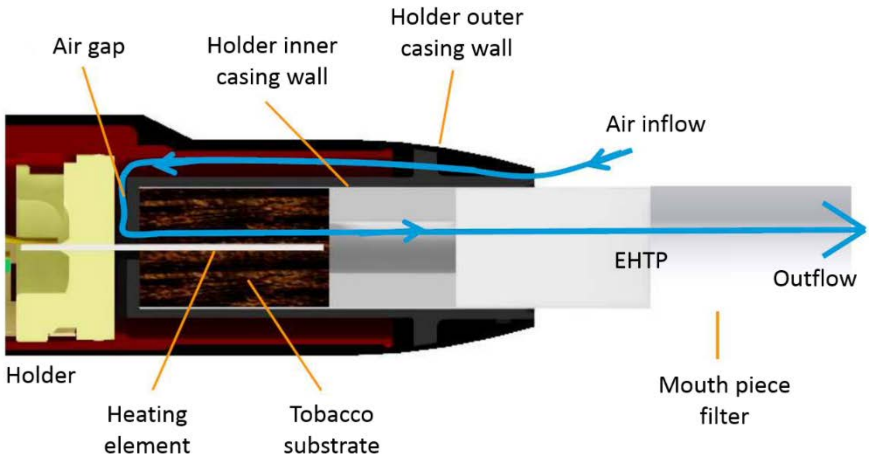
- **Charger** that is used to recharge the Holder after each use

- **EHTP** is a novel patent pending tobacco product with unique processed tobacco made from tobacco powder

- **Holder** into which the EHTP is inserted and which heats the tobacco material by means of an electronically controlled heater

How does an EHTP function?

- During use, the patented EHTS heats a specifically designed tobacco product (EHTP) at a controlled temperature when inserted into the Holder (heating device).
- The tobacco material in the EHTP undergoes a controlled heating process to release chemical compounds into gas phase, from which an aerosol is formed during cooling



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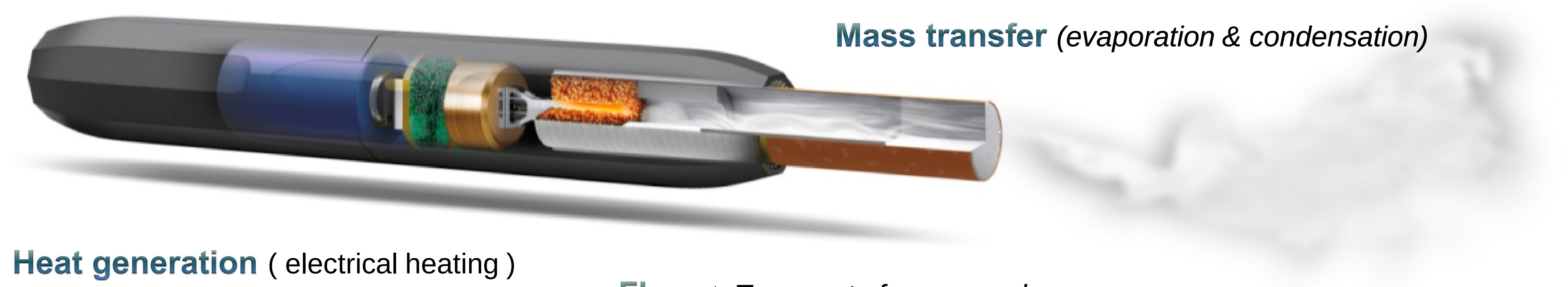
Physical and chemical processes governing aerosol generation in an EHTP

Release of Volatile Organic Compounds (VOCs) from tobacco

Mass transfer *(evaporation & condensation)*



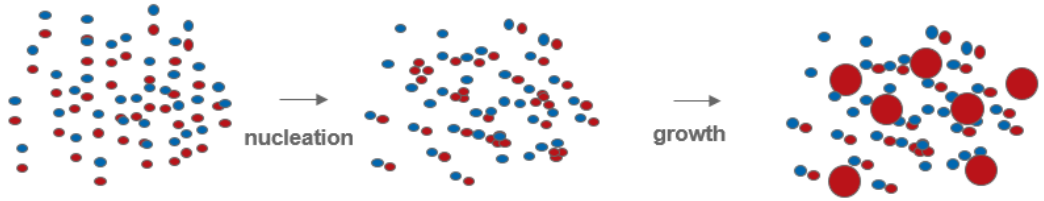
Heat generation (electrical heating)

Flow ⇨ *Transport of compounds*

Heat transfer (conduction, convection, radiation)

Aerosol formation & dynamics

Droplet size distribution, filtration & aerosol density,
Multicomponent gas and particulate phases



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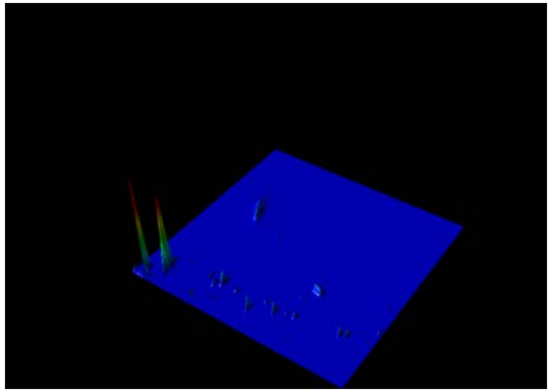
EHTP aerosol versus cigarette smoke

Constituents in the aerosol from PMI's EHTP (Health Canada regime)

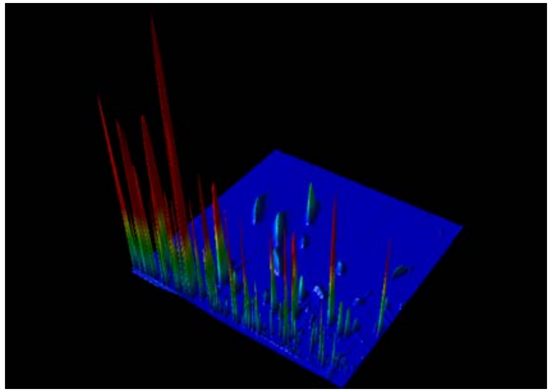


Resorcinol
Styrene
Catechol
Quinoline
Phenol
Pyridine
p-cresol
Toluene
Water
m-cresol
Benzene
o-cresol
Acrylonitrile
Formaldehyde
Isoprene
Air
1,3-Butadiene
Glycerol
Acetaldehyde
Nitrogen dioxide
Acetone
Nitric oxide
Acrolein
Carbon monoxide
Nicotine
Propionaldehyde
Crotonaldehyde
Methyl Ethyl Ketone
Butyraldehyde

Selected compounds quantified in the aerosol from the EHTP, identified as harmful, potentially harmful, or major constituents

Heating instead of burning reduces constituents

Constituents in cigarette smoke (3R4F, Health Canada regime)



Styrene
Quinoline
Resorcinol
Pyridine
Catechol
p-cresol
Toluene
Water
Phenol
o-cresol
m-cresol
Benzene
Acetaldehyde
Formaldehyde
Acrylonitrile
Acrolein
Acetone
Propionaldehyde
Isoprene
Air
Glycerol
1,3-Butadiene
Crotonaldehyde
Methyl Ethyl Ketone
Nitrogen dioxide
Nicotine
+ many additional compounds
Nitric oxide
Carbon monoxide
Butyraldehyde

[NTDS GC × GC-TOFMS: Volatile method]

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Objective

Investigate the aerosol formation characteristics for realistic operating conditions of the EHTS as well as for relevant gas mixture compositions measured in the EHTP aerosol

- Determine from which compounds and at what operating conditions aerosol droplets are formed when the tobacco in the EHTP is heated.
- The role of compounds resulting from evaporation, such as water, glycerol and nicotine, on the aerosol formation
- Evaluate the potential for aerosol droplet nuclei formation from thermal degradation products alone, in the absence of any aerosol former

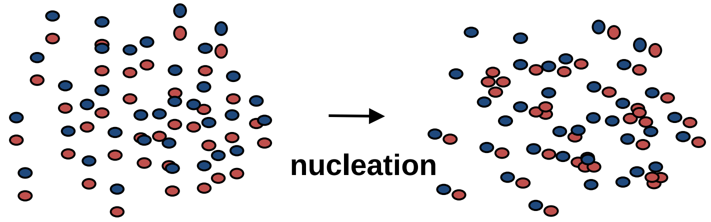
MODELING AEROSOL FORMATION

Modeling aerosol formation in an EHTP

- Computational Fluid Dynamics (CFD) with heat, mass and species transfer and aerosol formation and dynamics
- An extended Classical Nucleation Theory (CNT) for multicomponent gas mixtures
- Eulerian-Eulerian treatment of both gas and liquid (droplet) phases *(both phases are treated as continua)*

Mass conservation	$\partial_t \rho + \partial_j(\rho u_j) = 0$
Momentum conservation	$\partial_t(\rho u_i) + \partial_j(\rho u_i u_j) = -\partial_i p + \partial_j \tau_{ij} \quad ; \quad i = 1, ..., 3$
Energy conservation	$\rho c_p \left(\partial_t T + u_j \partial_j T\right) = -\partial_j q_j - \tau_{kj}\partial_k u_j + S_h + \frac{Dp}{Dt}$
Transport of compounds in gas phase	$\partial_t(\rho Y_i) + \partial_j(\rho Y_i u_j) = -\partial_j j_{j,i} + S_i^{l \to v} \quad ; \quad i = 1, ..., n$
Transport of compounds in droplet phase	$\partial_t(\rho Z_i) + \partial_j(\rho Z_i u_j) = -S_i^{l \to v} \quad ; \quad i = 1, ..., n$
Transport of droplet number density	$\partial_t(N) + \partial_j(N u_j) = J_N - J_c - J_{ev}$

Homogeneous nucleation according to the extended CNT



Supersaturated vapors with carrier gas

Temperature

Nucleation rate:

$$\boxed{J_N = R_{av} Z c_{eq}}$$

Zeldovich factor

$$c_{eq} = \exp\left(-\frac{\Delta G}{kT}\right) \prod_{i=1}^{n} \left(\frac{p_i^{\mathrm{sat}}(T)}{kT} \exp\left(\frac{s_i^{\mathrm{mon}} \sigma_i}{kT}\right)\right)^{w_i}$$

Gibbs free energy

Saturation vapor pressure

'monomer' surface area

surface tension

mole fraction

Boltzmann's constant

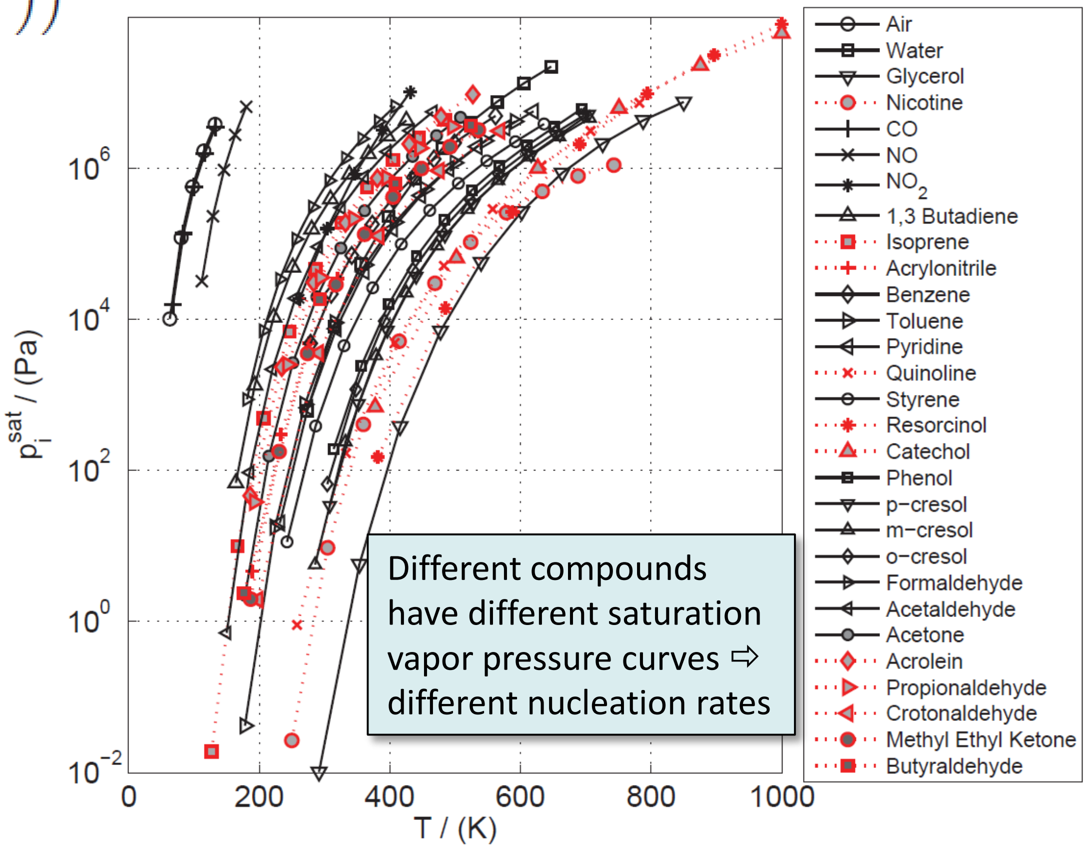
Temperature

$$R_{av} = \frac{\sum_{i=1}^{n} N_i^2}{\sum_{i=1}^{n} N_i^2 / K_{ii}}$$

Average growth rate

condensation rate

number of molecules of component *i* in a critical (equilibrium) cluster

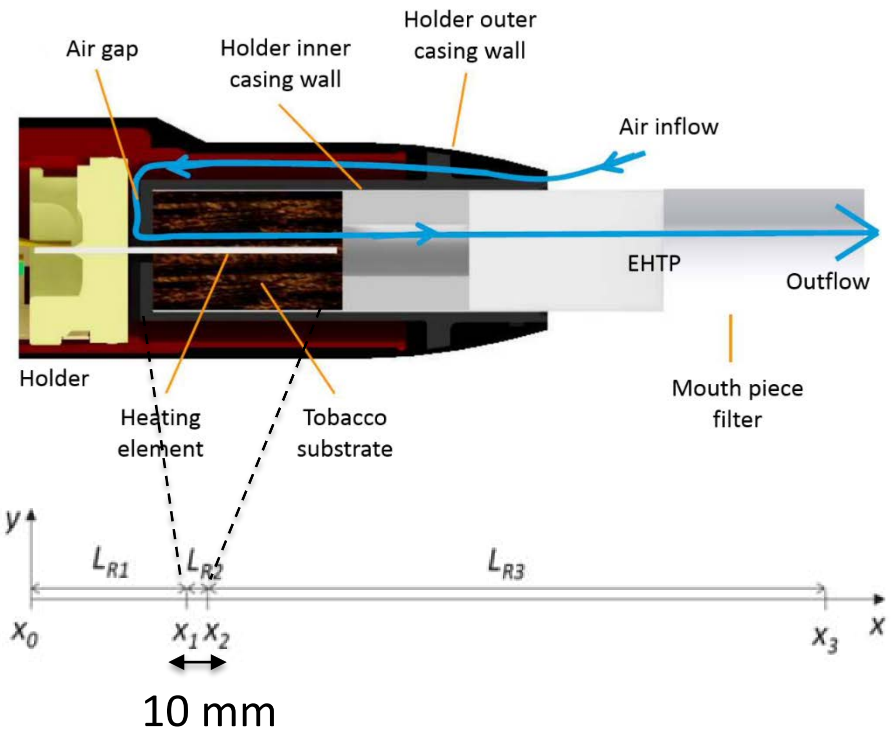


SIMULATION CASES

Simulation geometry and estimated operating conditions

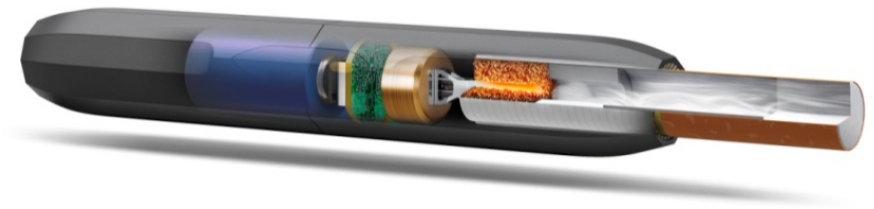
Simulation geometry

- One-dimensional with representative dimensions of geometric dimensions and cooling rates along the flow path



Estimated operating conditions

- Temperature range 30°C – 350°C *(controlled by electrical heater)*
- Velocity range in the product estimated from a Health Canada Intense puff profile *(55mL puff during 2 seconds, every 30 seconds)*
- Estimated cooling rates in the EHTP:
  - Low cooling rate: LCR = $-2.2\cdot10^4$ K/s
  - High cooling rate: HCR = $-2.9\cdot10^5$ K/s



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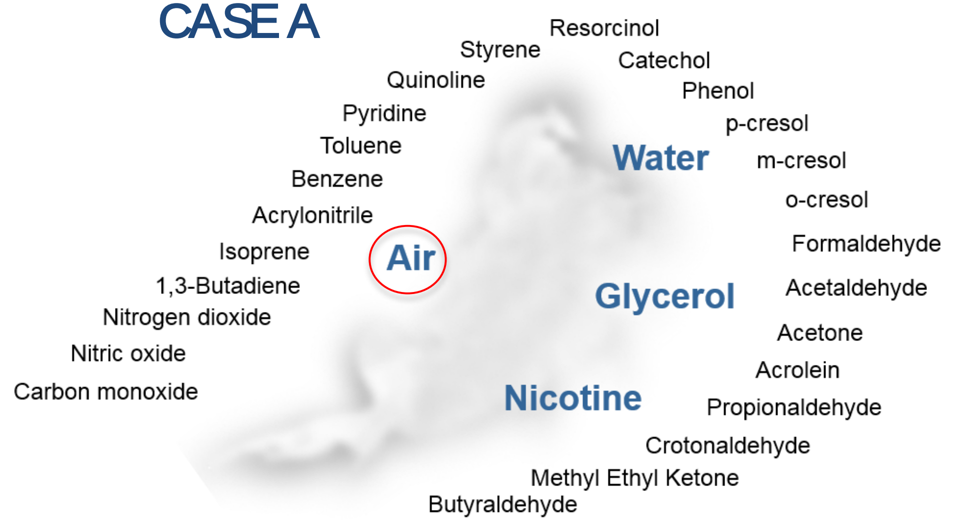
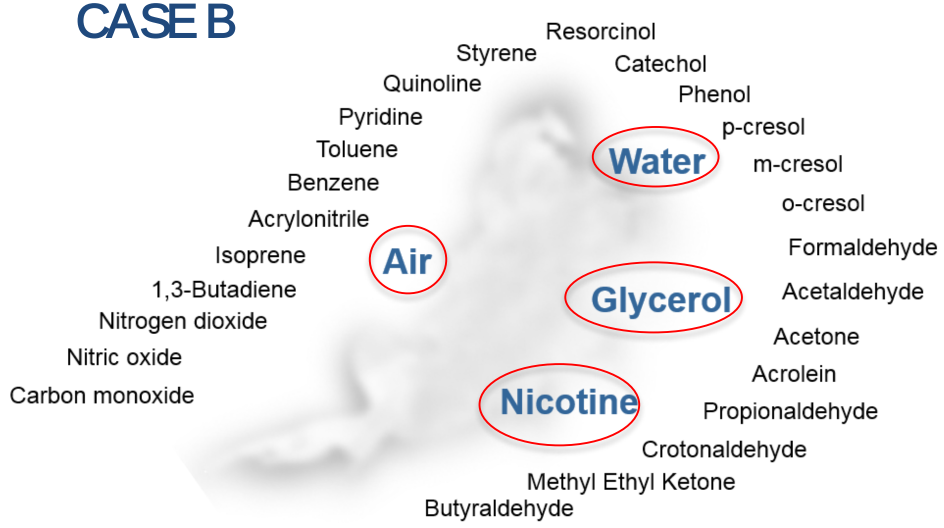
Chemical composition in cases

CASE A



- All compounds except for air may contribute to the formation of aerosol droplets
- Masses assumed to be evenly distributed over all puffs and initially in the gas phase

CASE B



- Same as case A, but compounds originating from the tobacco by evaporation, are treated as inert and cannot contribute to the aerosol droplet formation.

Investigate if compounds generated from thermal decomposition of tobacco components can form aerosol droplets on their own, without the help of aerosol formers released from the tobacco by evaporation.

Compound present in multicomponent gas mixture, but cannot contribute to the formation of aerosol droplets

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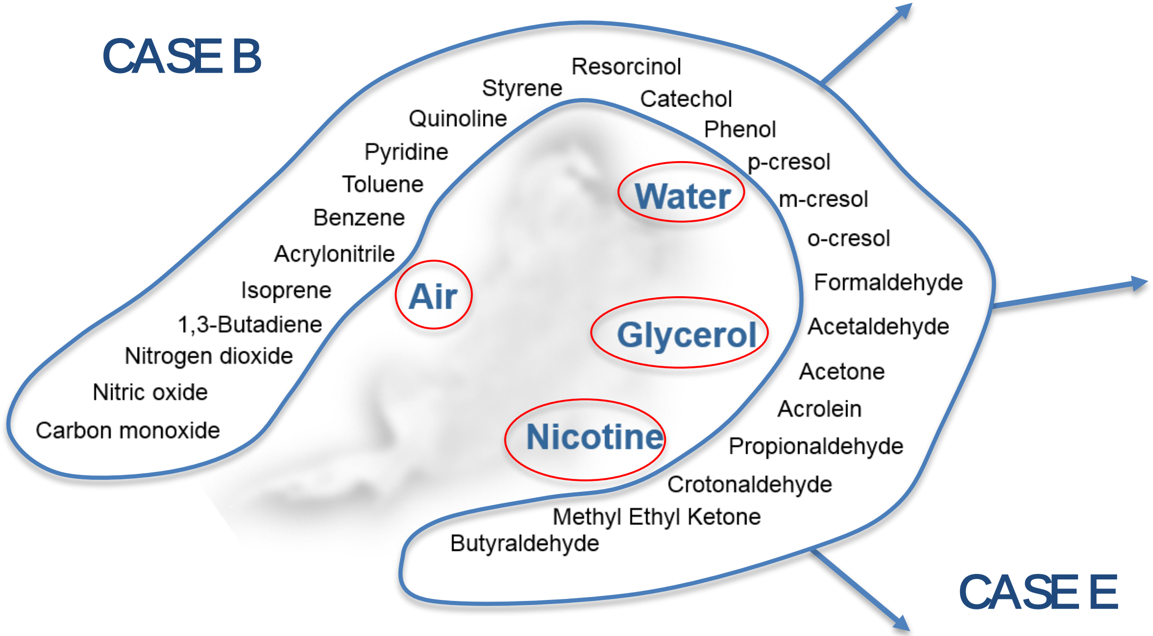
Chemical composition in cases

CASE C

$\times 50$

- Represents ~1.5 times the total mass remainder reported by Labstat International ULC and omitted in the simulation cases A and B.

CASE B



CASE D

$\times 100$

- Represents a situation where all the mass of the minor components appears during roughly 1/8 of the duration of a single puff of 2s.
- Collected mass of the minor compounds is almost equal to the mass of water

CASE E

$\times 500$

- Extreme case to investigate the sensitivity of the nucleation process and to estimate the likelihood of aerosol droplets being generated from the minor components without the help of an aerosol former.

Compound present in multicomponent gas mixture, but cannot contribute to the formation of aerosol droplets

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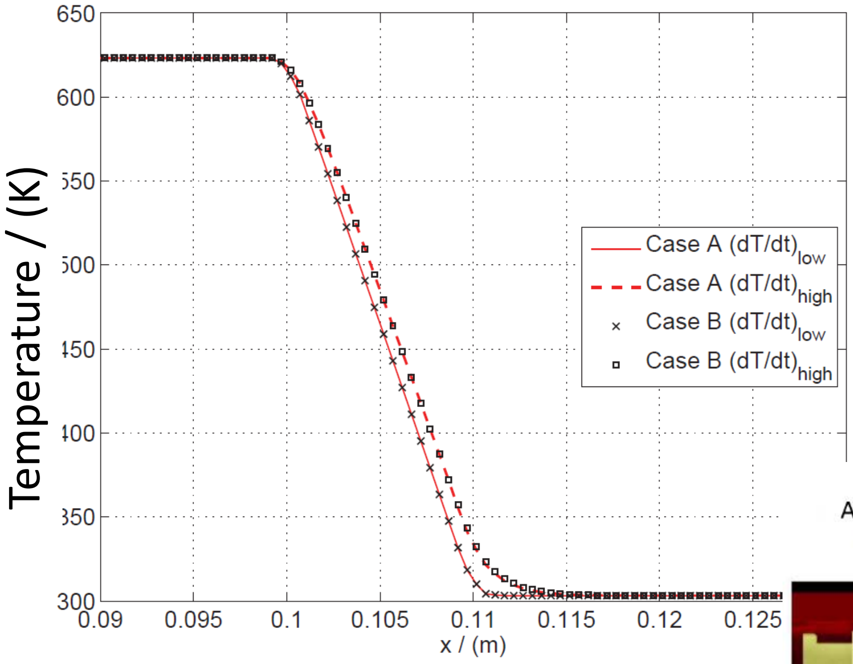
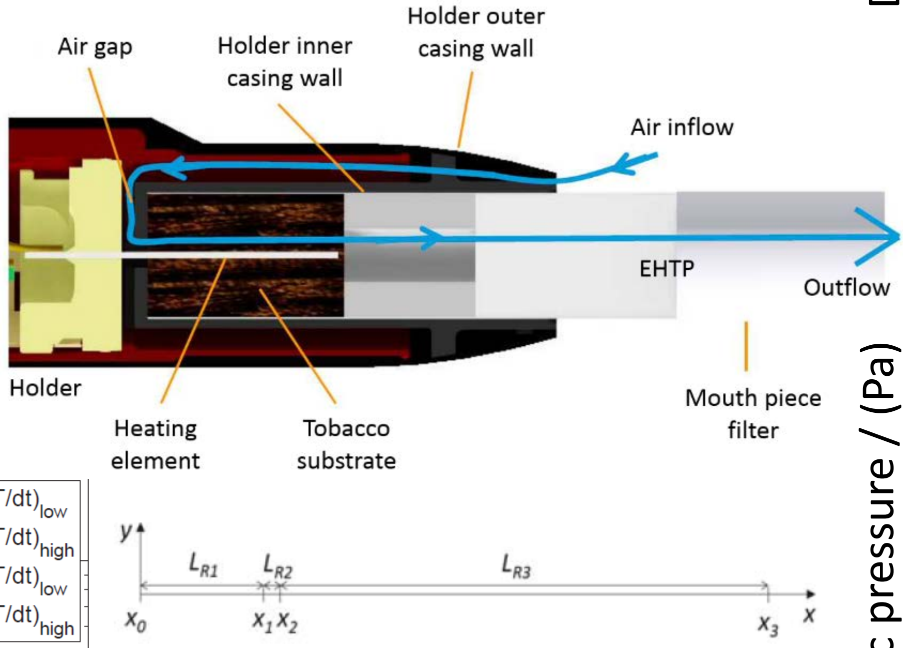
RESULTS

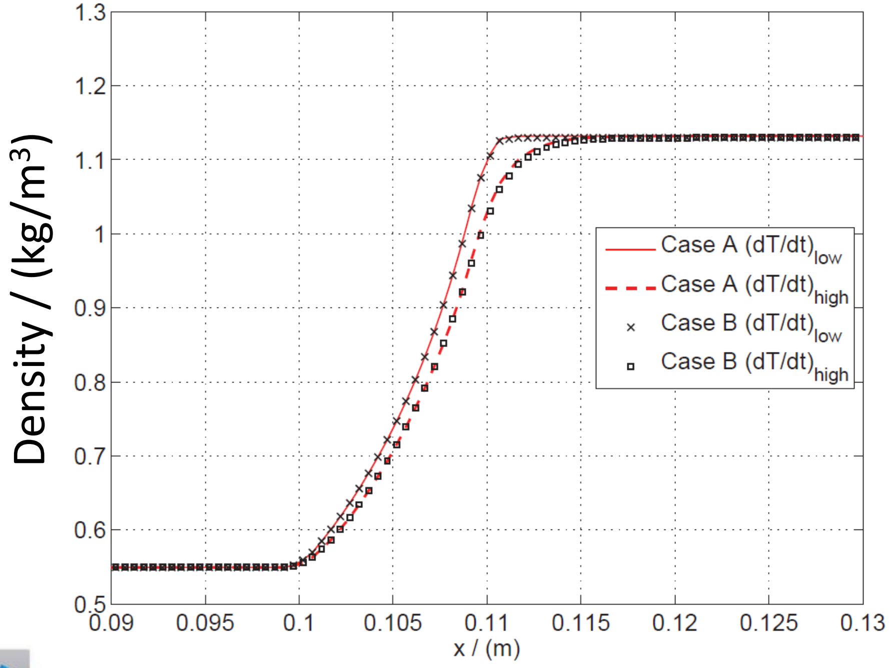
Cases A & B



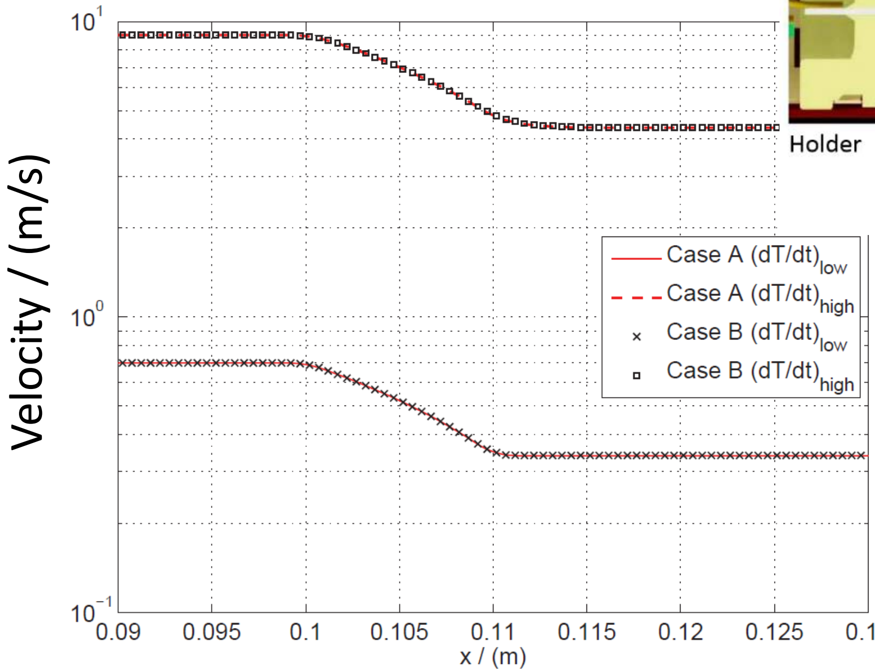
Temperature / (K)
x / (m)
Case A (dT/dt)low
Case A (dT/dt)high
Case B (dT/dt)low
Case B (dT/dt)high



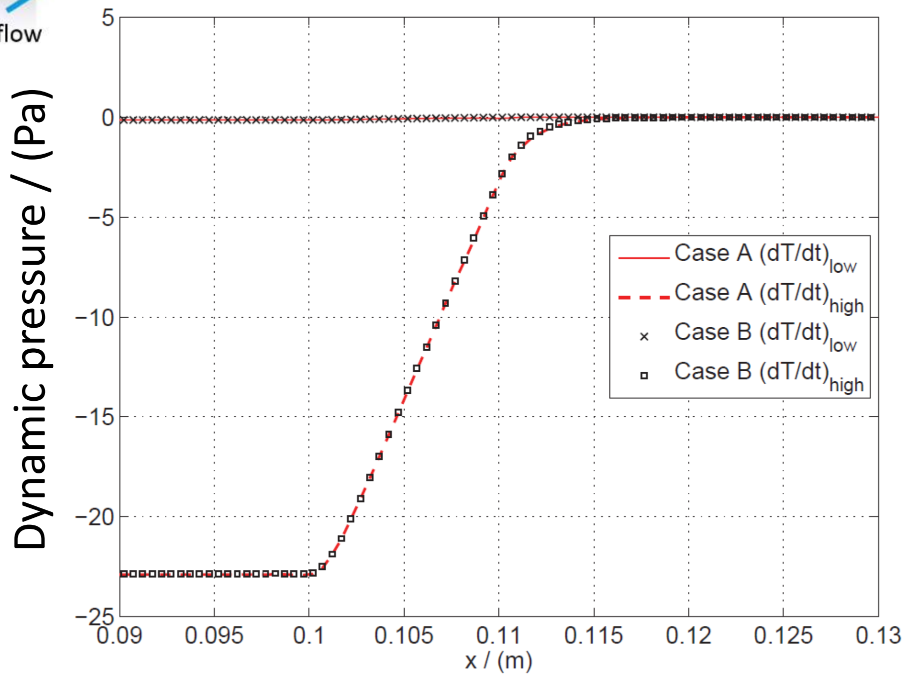
Air gap
Holder inner casing wall
Holder outer casing wall
Air inflow
EHTP
Outflow
Holder
Heating element
Tobacco substrate
Mouth piece filter
y
L_{R1}
L_{R2}
L_{R3}
x_0
x_1 x_2
x_3
x



Density / (kg/m^3)
x / (m)
Case A (dT/dt)low
Case A (dT/dt)high
Case B (dT/dt)low
Case B (dT/dt)high

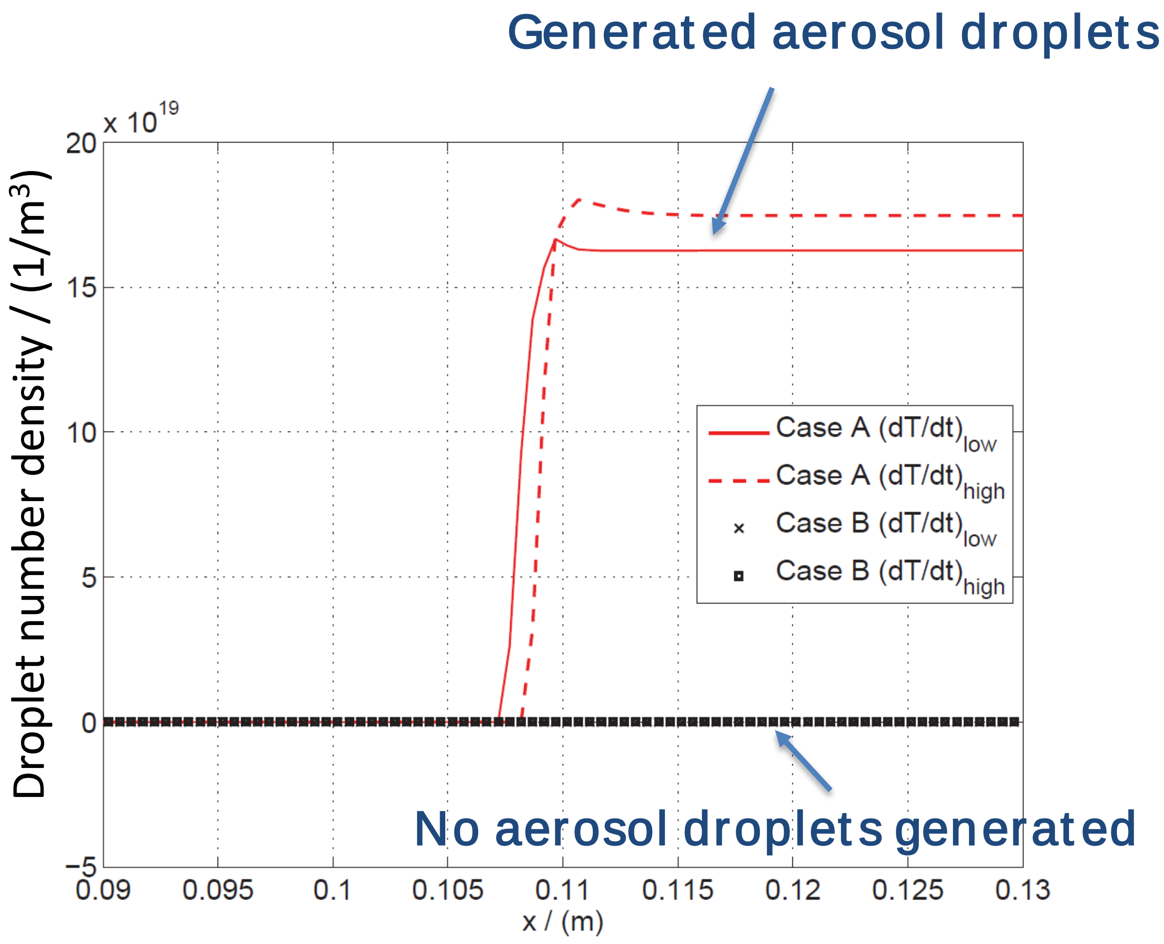


Velocity / (m/s)
x / (m)
Case A (dT/dt)low
Case A (dT/dt)high
Case B (dT/dt)low
Case B (dT/dt)high



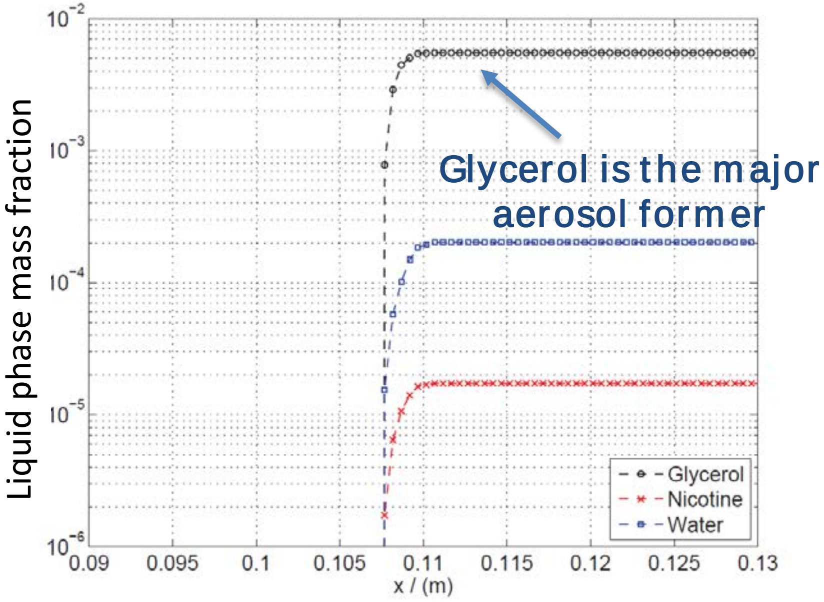
Dynamic pressure / (Pa)
x / (m)
Case A (dT/dt)low
Case A (dT/dt)high
Case B (dT/dt)low
Case B (dT/dt)high

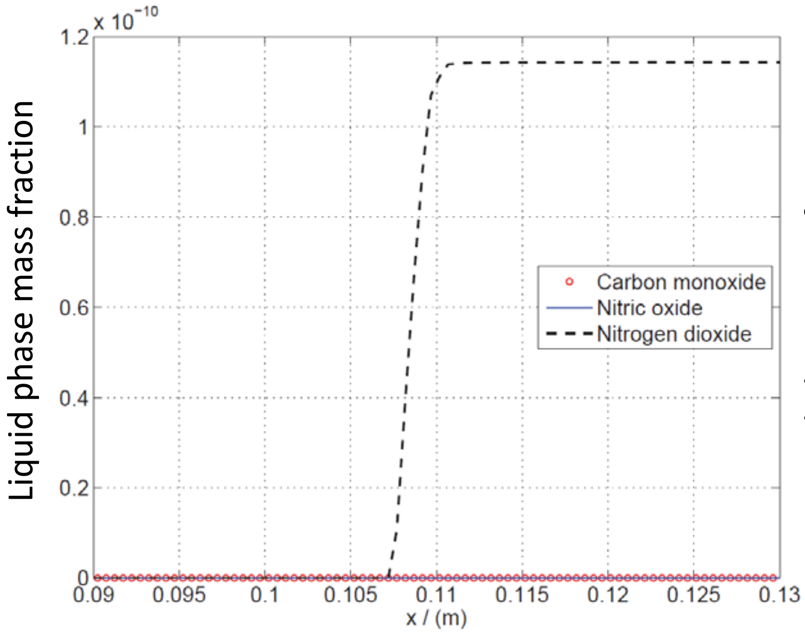
Aerosol formation in cases A & B

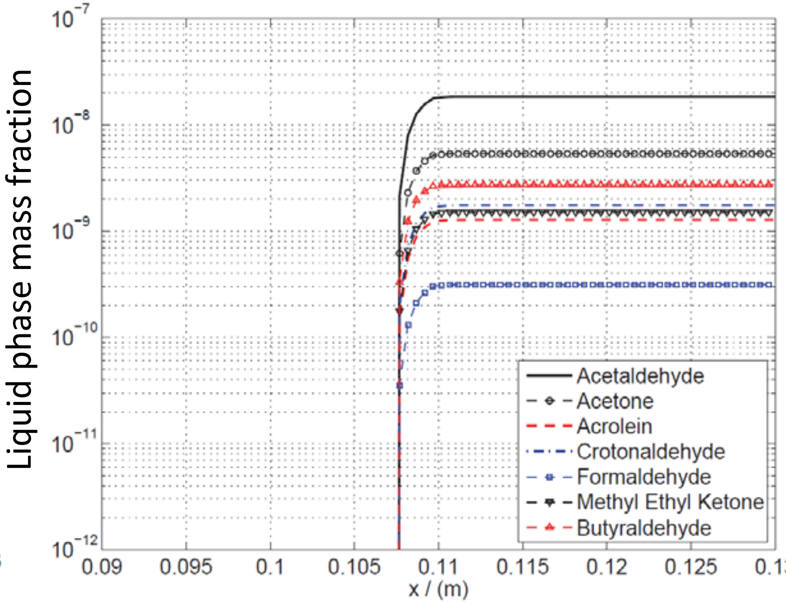


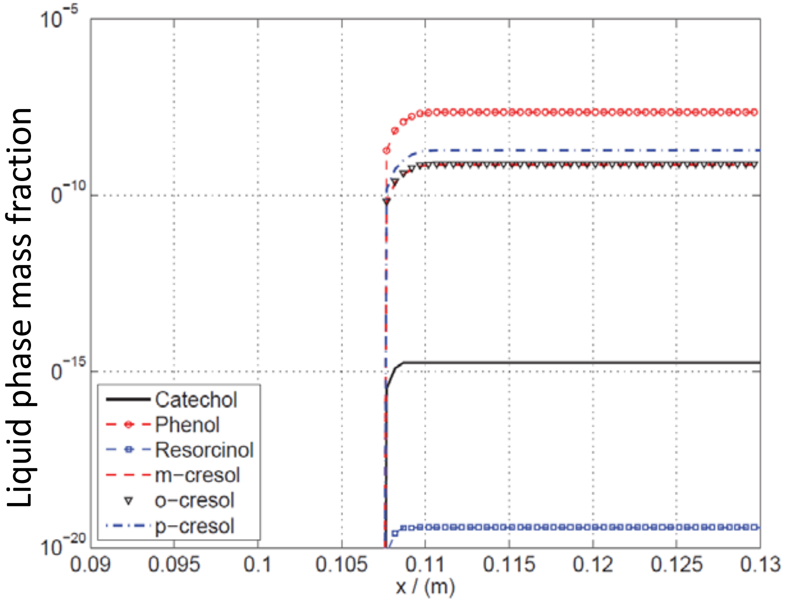
- Aerosol droplet nuclei are only generated in case A
- Only minor influence of cooling rate on nucleation in the EHTP
- An aerosol former (glycerol) is required to generate aerosol droplets

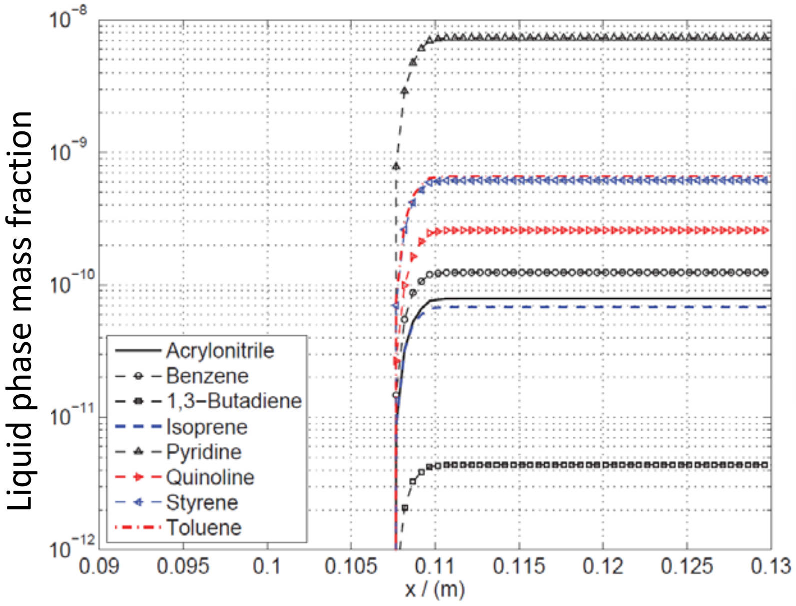
Liquid mass fractions in nuclei generated in case A











- Glycerol is the major aerosol former
- Only minor liquid mass fraction of other compounds in the nuclei generated by the glycerol

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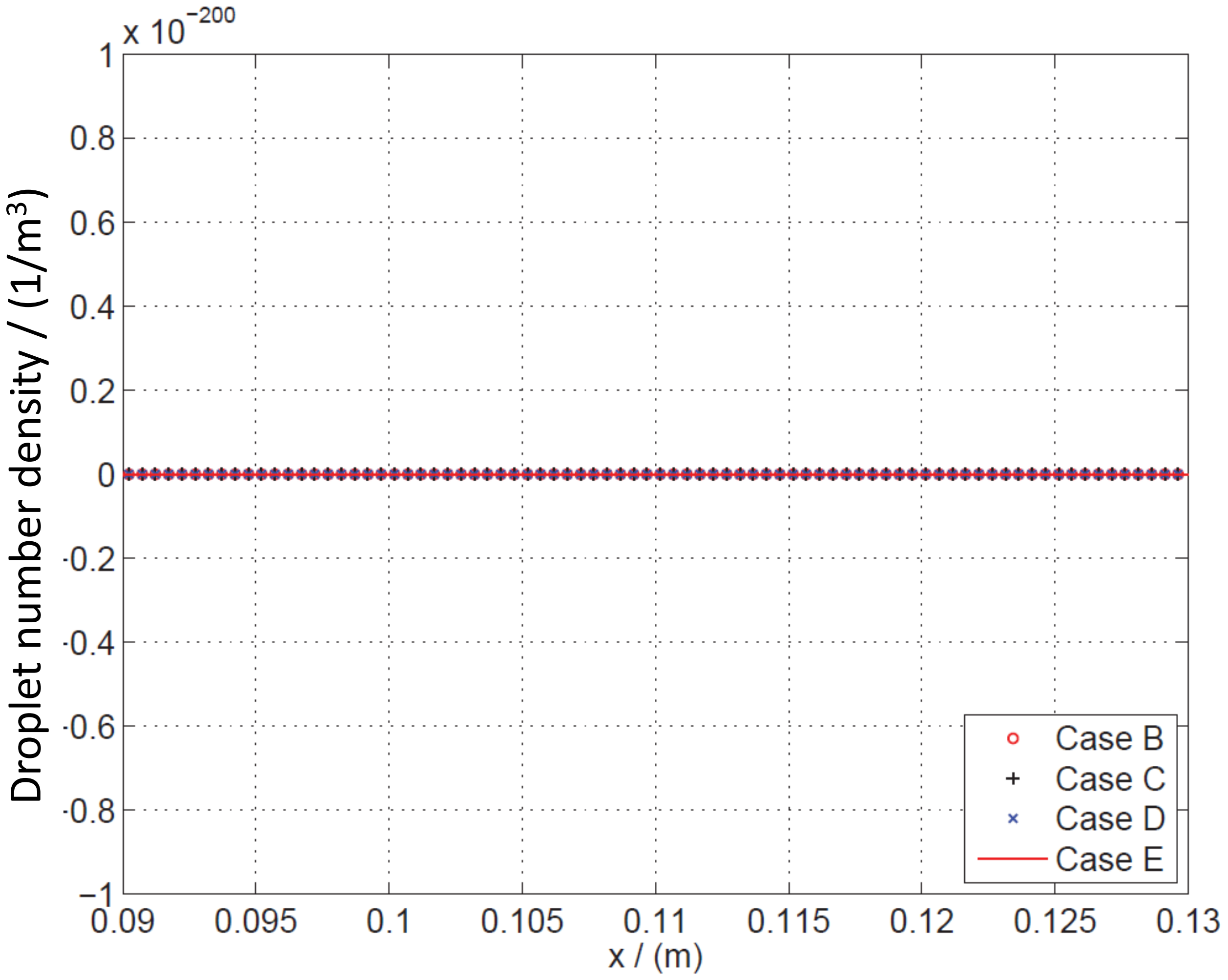
Aerosol formation in cases C-E



According to the extended CNT, aerosol droplet nuclei cannot form from chemical compounds potentially derived from the thermal degradation of tobacco components during use of the EHTP

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CONCLUSIONS

Conclusions

- Aerosol formation in the EHTP during use when heated in the Holder (heating device) has been modeled using an extended CNT for multicomponent gas mixtures *to investigate the aerosol formation characteristics for realistic heating operating conditions of the EHTP as well as for relevant gas mixture compositions of the EHTP aerosol*

- Simulations show that aerosol droplets are formed only by the activity of an aerosol former being mainly glycerol *(evaporated from the tobacco plug when it is used as intended and heated by the Holder)*

- According to the extended CNT for multicomponent aerosols, an aerosol cannot be generated from the minor compounds of the gas mixture alone at the compositions and operating conditions tested, *i.e. representative of EHTP use conditions, even if all minor compounds, potentially generated via thermal decomposition reactions, are released or generated in a single puff*

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