Nanoparticle formation in Diesel vehicle exhaust: First measurements of precursor gases

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Abstract

Nano particles generated by Diesel vehicles are of considerable current concern since they represent major air pollutants in cities, near motor ways, and in certain work places. Modern Diesel exhaust after treatment systems remove engine generated soot and ash particles and organics, but promote the formation of numerous nucleation particles (NUP) formed in the rapidly cooling exhaust by nucleation of low vapour pressure exhaust gases. NUP have diameters around 10 nm, which is exactly the diameter range allowing most efficient aerosol particle intrusion into the lowest, least protected, and most vulnerable compartment of the human lung. However, the NUP nature and mechanism of formation are only poorly explored. Using a novel mass spectrometric method, we have made the first systematic on line measurements of acidic NUP precursor gases in Diesel exhaust. The experiments employed a modern heavy duty Diesel vehicle engine operated with and without exhaust after treatment and combusting low sulphur fuels and bio fuel. Measured exhaust gases include the key NUP precursor sulphuric acid and other acidic gases including di-sulphuric acid, and low vapour pressure di-carboxylic acids. In addition we have made simultaneous NUP measurements and NUP model simulations. We find that modern Diesel exhaust after-treatment systems promote NUP formation by mediating the formation of nucleating and condensing gases.

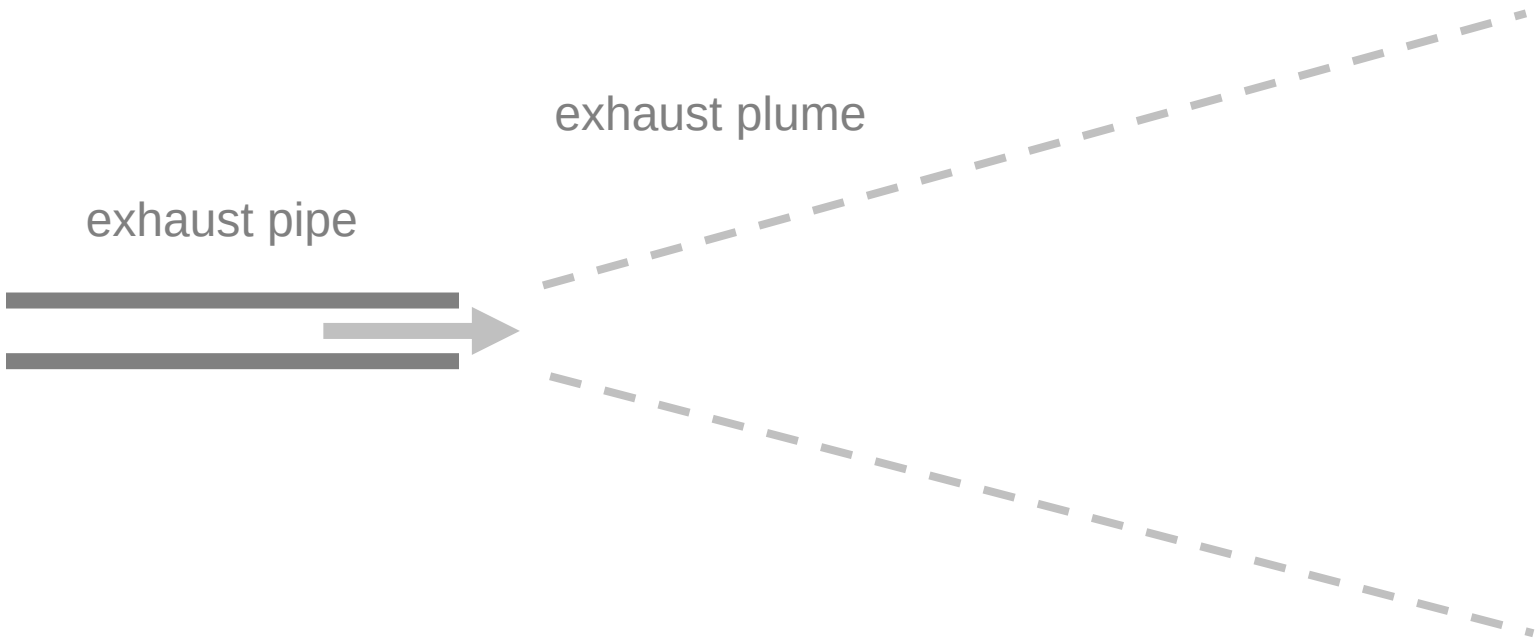
Diesel Nano Particles :

- major air pollutants in cities, near motorways, and in certain work places
- potential for inducing adverse health effects

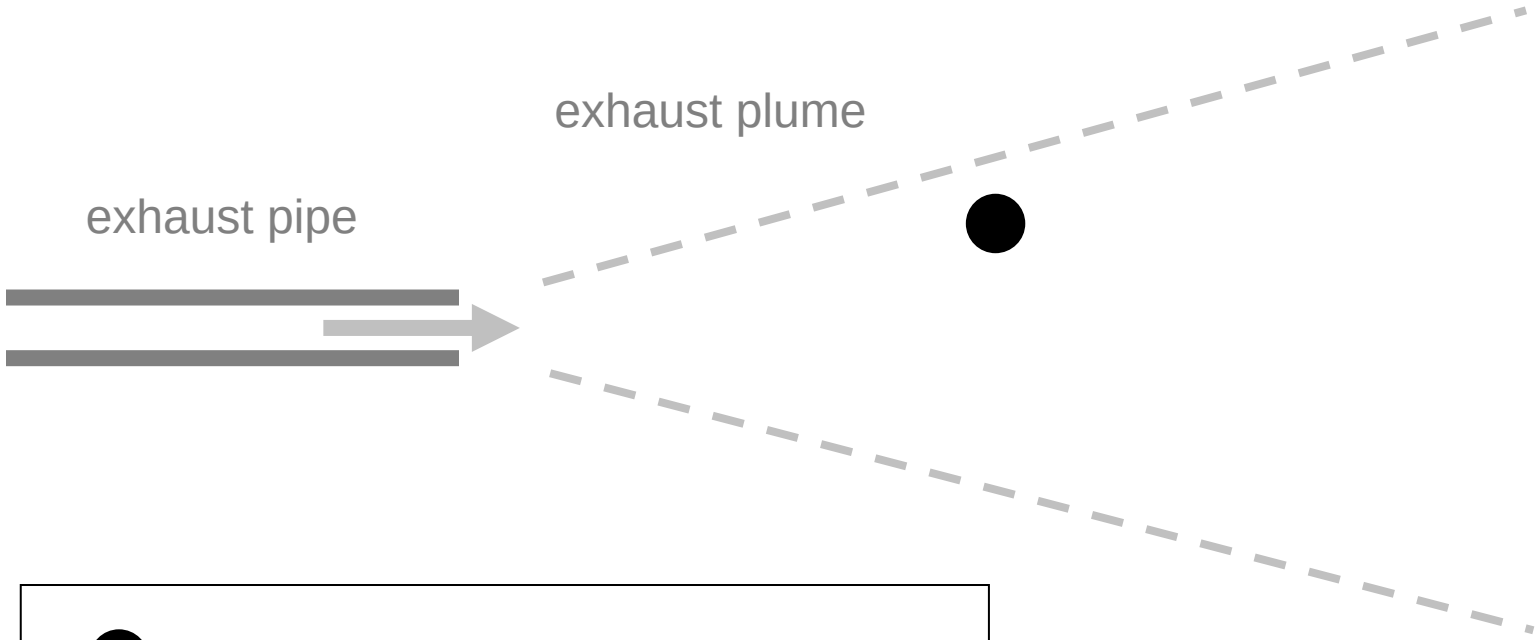
Diesel Nano Particles :

2 families

Diesel exhaust : **without** after treatment

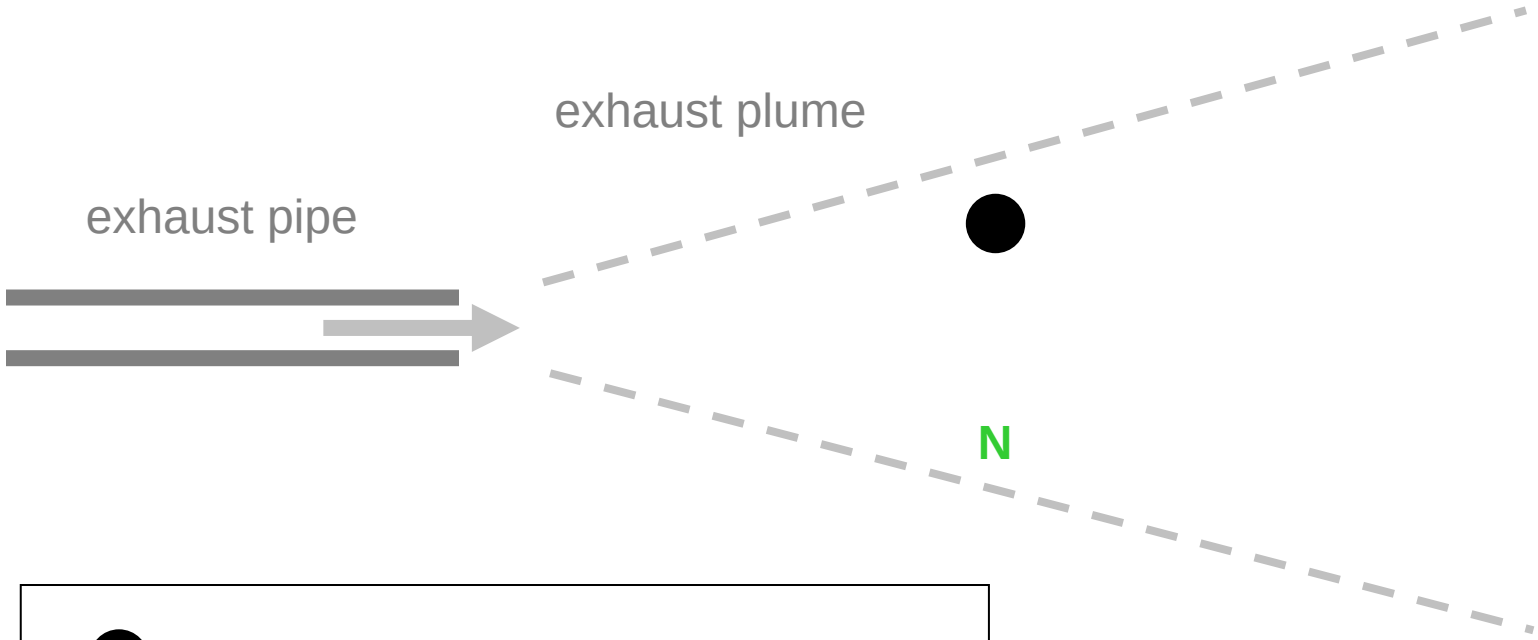


Diesel exhaust : **without** after treatment



Primary particle (soot , D=40-100 nm)

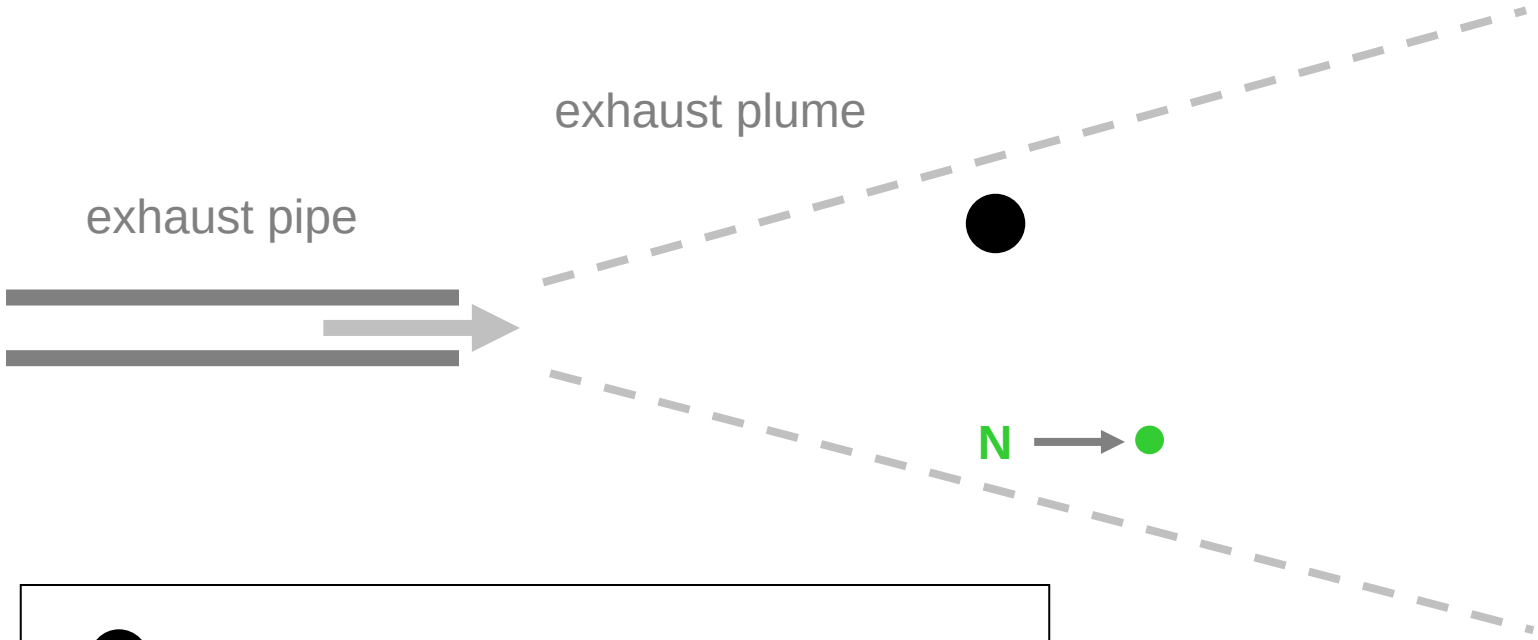
Diesel exhaust : without after treatment



Primary particle (soot , D=40-100 nm)

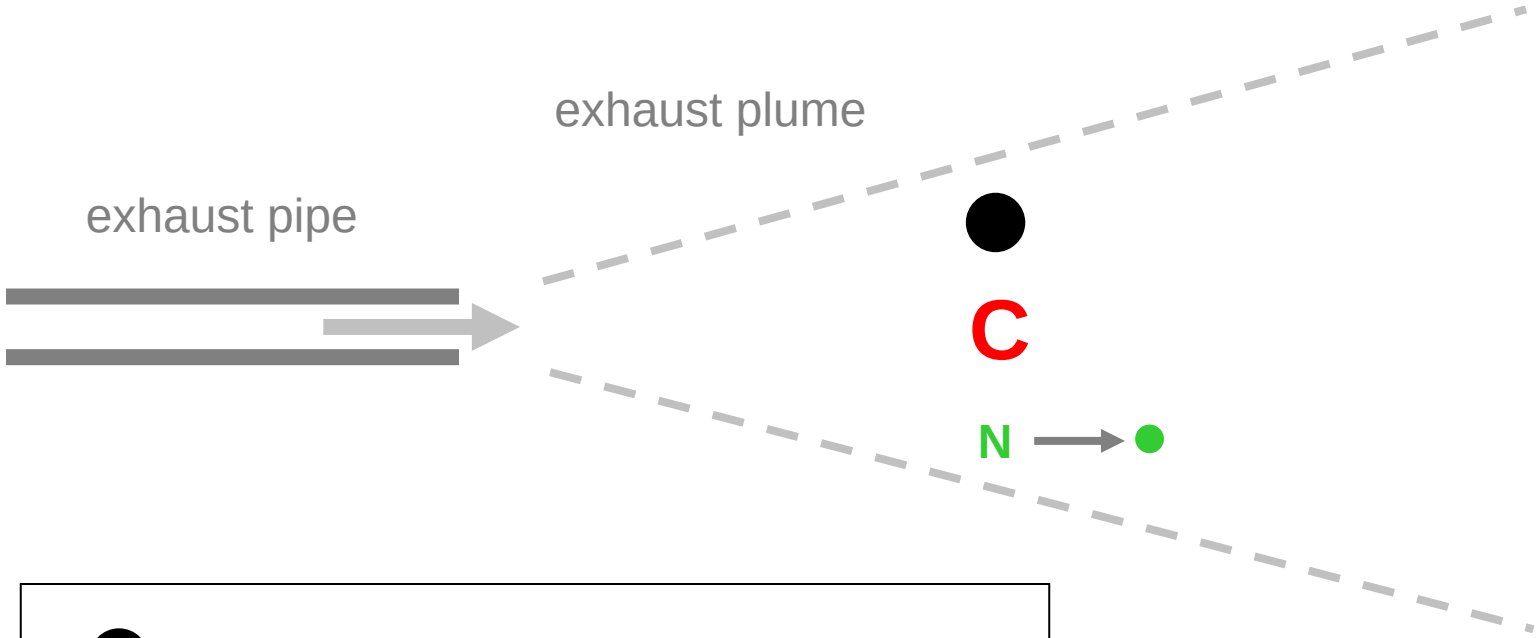
N nucleating gas molecule (H_2SO_4)

Diesel exhaust : without after treatment



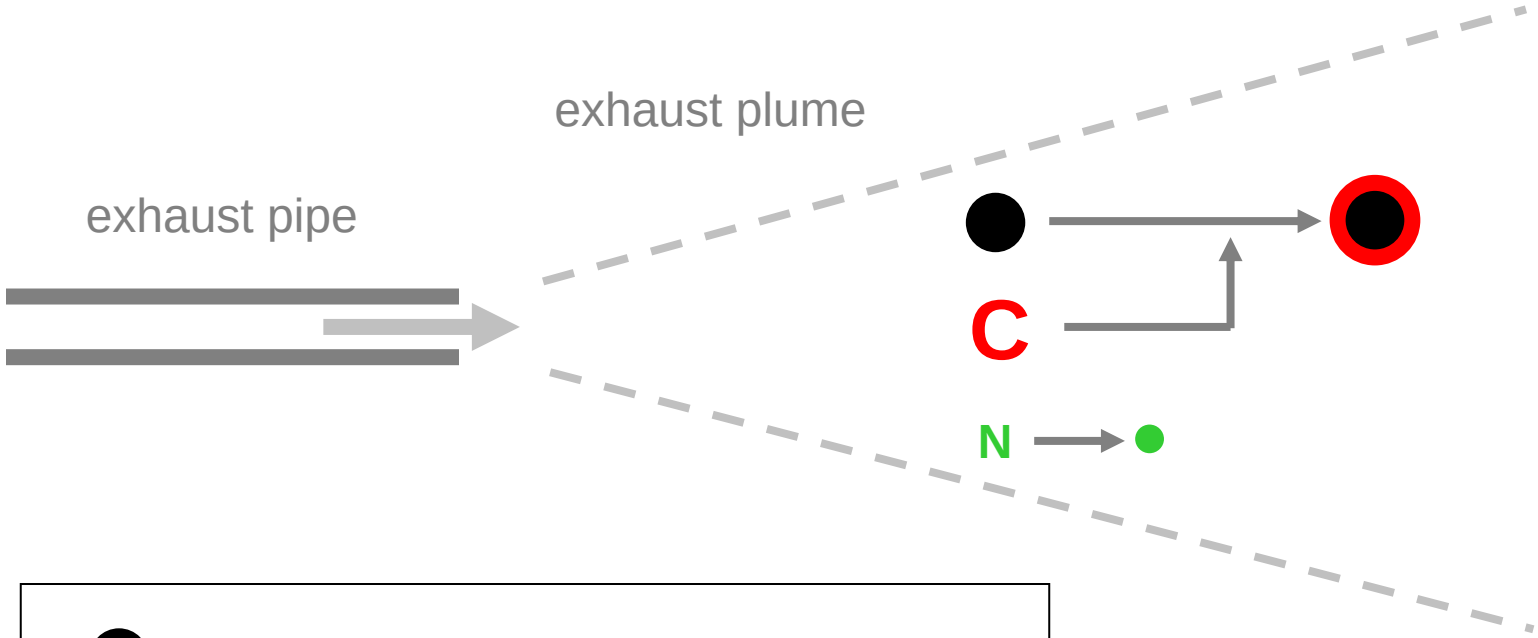
●	**Primary particle (soot , D=40-100 nm)**
N	**nucleating gas molecule (H2SO4)**
●	**Nucleation mode particle NMP (D=1nm)**

Diesel exhaust : without after treatment



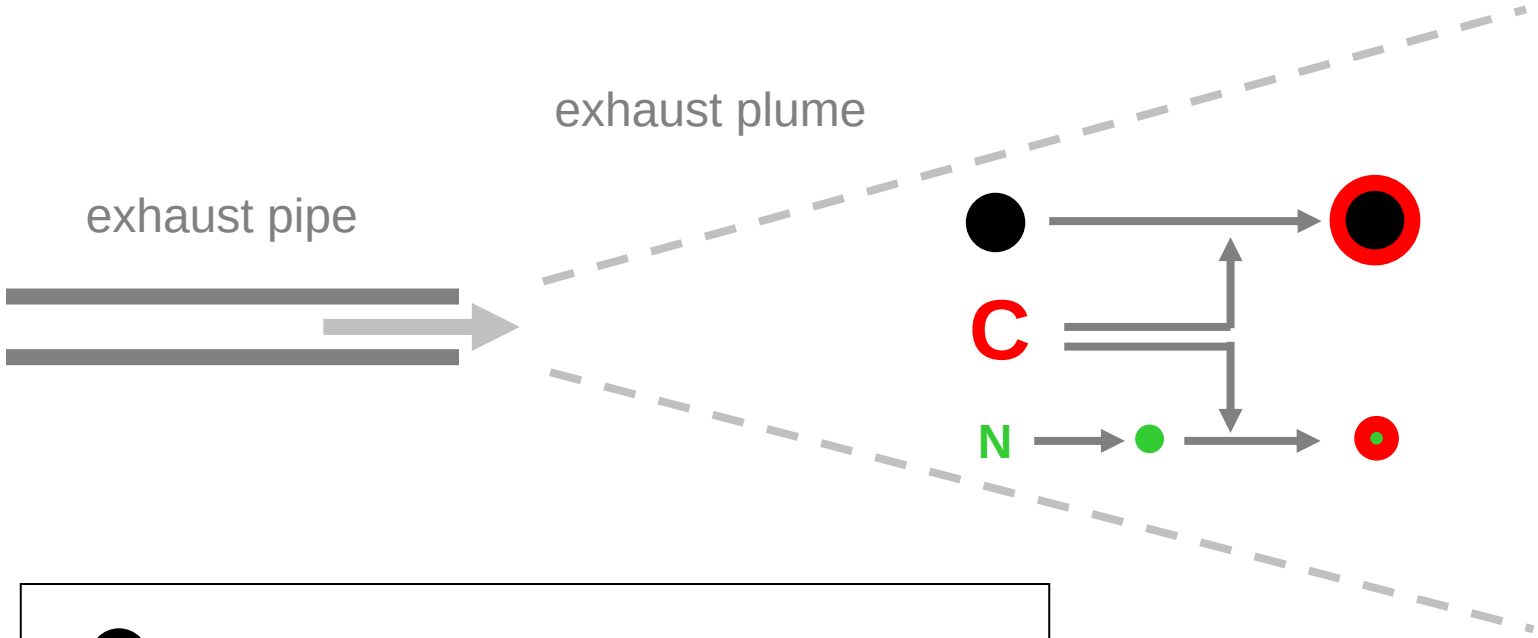
●	**Primary particle (soot , D=40-100 nm)**
C	**condensing gas molecule**
N	**nucleating gas molecule (H2SO4)**
●	**Nucleation mode particle NMP (D=1nm)**

Diesel exhaust : without after treatment



- Primary particle (soot , D=40-100 nm)
- C — condensing gas molecule
- N — nucleating gas molecule (H2SO4)
- Nucleation mode particle NMP (D=1nm)
- coated primary particle (soot) ; D=40-100 nm
- coated NMP (D=5-20 nm)

Diesel exhaust : **without** after treatment



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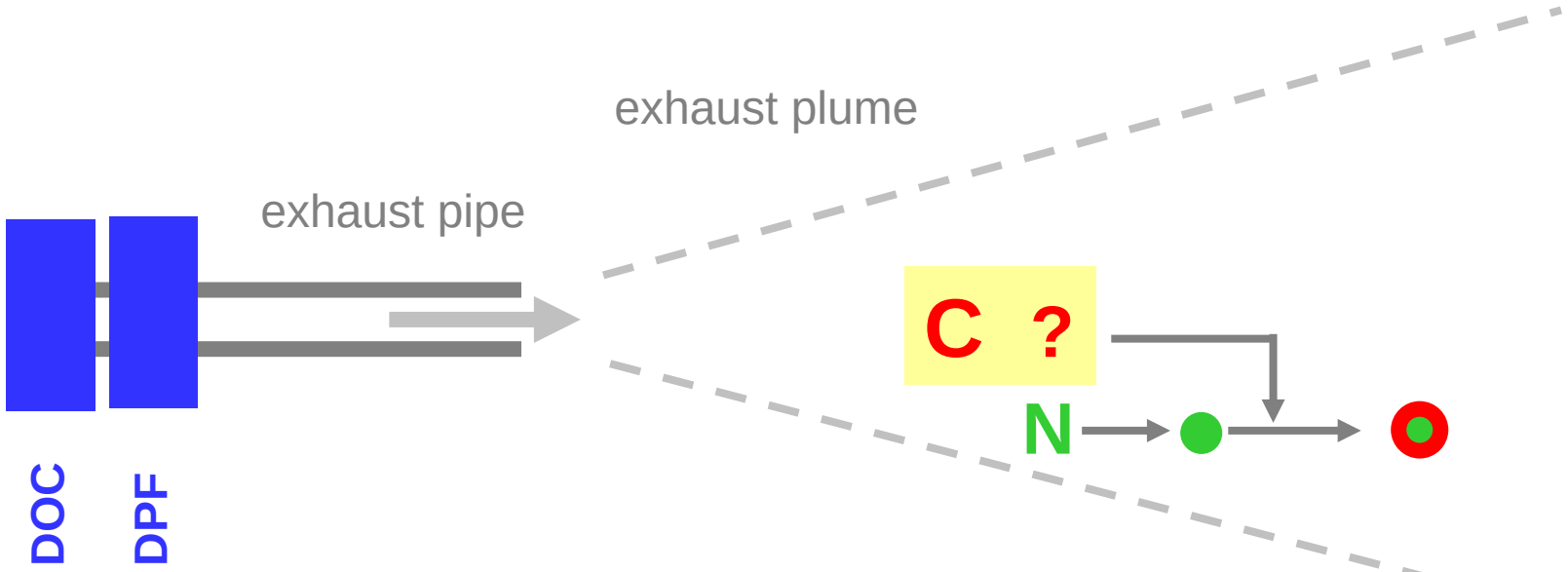
Diesel exhaust : with after treatment (DOC + DPF)



Nucleation particles (NUP)

- Mechanism of formation and chemical nature only poorly understood
- NUP precursor gases not known

Our plan

- Introduction of inovative measurement methods
- Measurements of NUP precursor gases in exhaust

Experiments
at MAN engine test lab (Nuernberg)

- **NUP precursors**: measurements in heated Diesel vehicle engine exhaust

  Measurement method: IMR-ITMS

  - developed by ***MPIK Heidelberg***

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    fragment ion analysis of mass selected ions

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  - ionization by ion-molecule reactions (IMR)
    soft and selective
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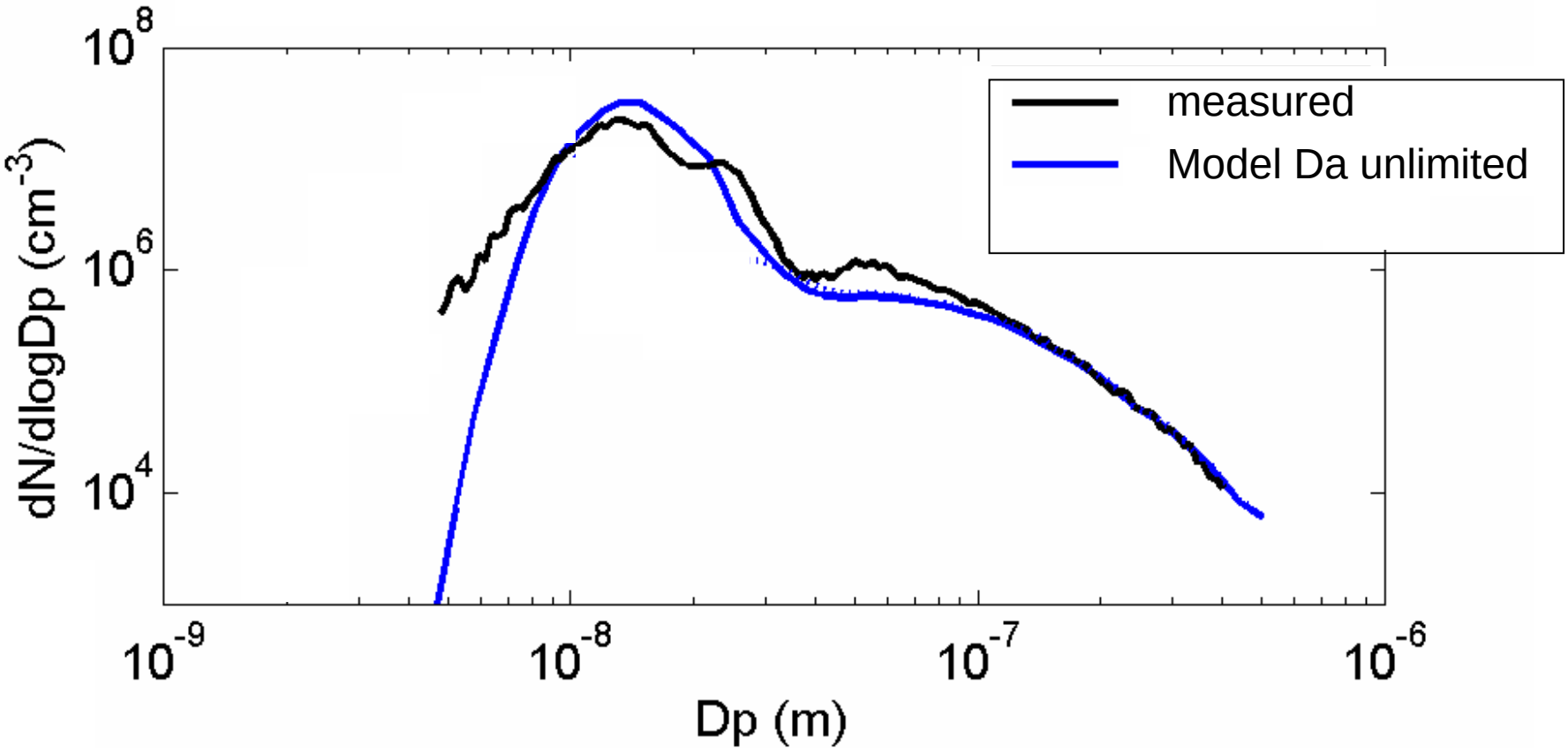
    fragment ion analysis of mass selected ions
    → greatly improves ion identification
- **NUPs**: formation and growth in flow tube (2.6 s)

Experimental conditions

- **Engine:**
  MAN, 440 PS, 6 cylinder turbo charge common rail EURO 4, 10.6 l, peak torque 2220 Nm
- **Fuels:**
  BIO (FSC: <1 ppmm)
  Standard (FSC: 6, 36 ppmm)
- **Engine loads:** 25, 30, 75, 100 %
- **Engine speed:** 1800 rpm
- **After treatment scenarios**
  - none
  - DOC+POC
  - DOC+DPF
  - DOC
  - DPF

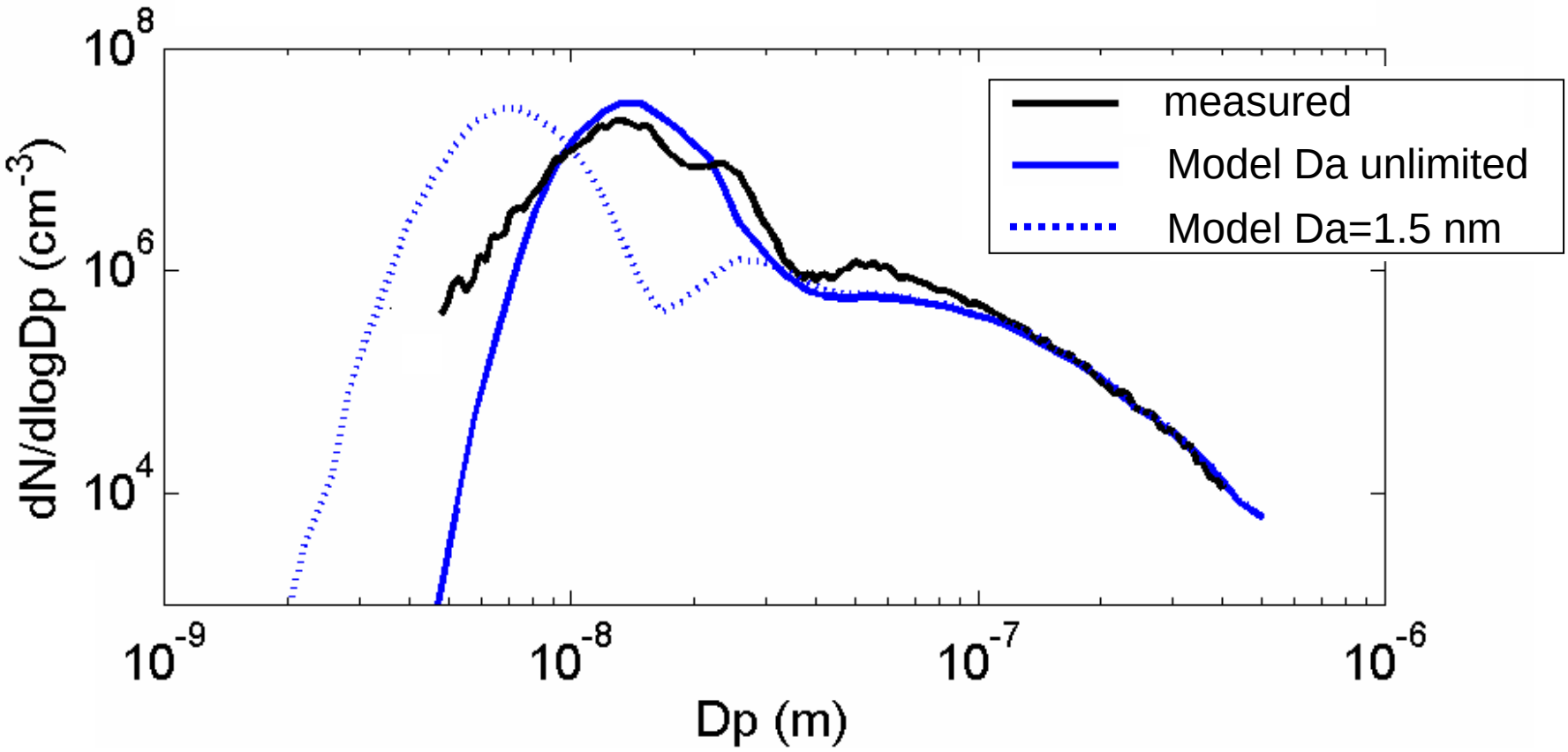
28 Nov 2007: DOC+POC(ECO) , FSC=36 ppmm , EL=100% ,



Measurement:Topi Rönkkö et al

Model: Liisa Pirjola

28 Nov 2007: DOC+POC(ECO) , FSC=36 ppmm , EL=100% ,



Measurement:Topi Rönkkö et al

Model: Liisa Pirjola

More figures

- Scientific publication in preparation
- For info contact:
  frank.arnold@mpi-hd.mpg.de

Conclusions

- Gaseous H2SO4: strong store and release effects
- NUP conc. increases with H2SO4
- NUP diameter increases with H2SO4
- NUP volume conc. Increases with H2SO4
- Acidic gases other than H2SO4 are also present
- …..

Thank You