
# REMOTE SENSING OF ATMOSPHERIC CONTENT IN BLACK CARBON, DUST AND GREENHOUSE GASES

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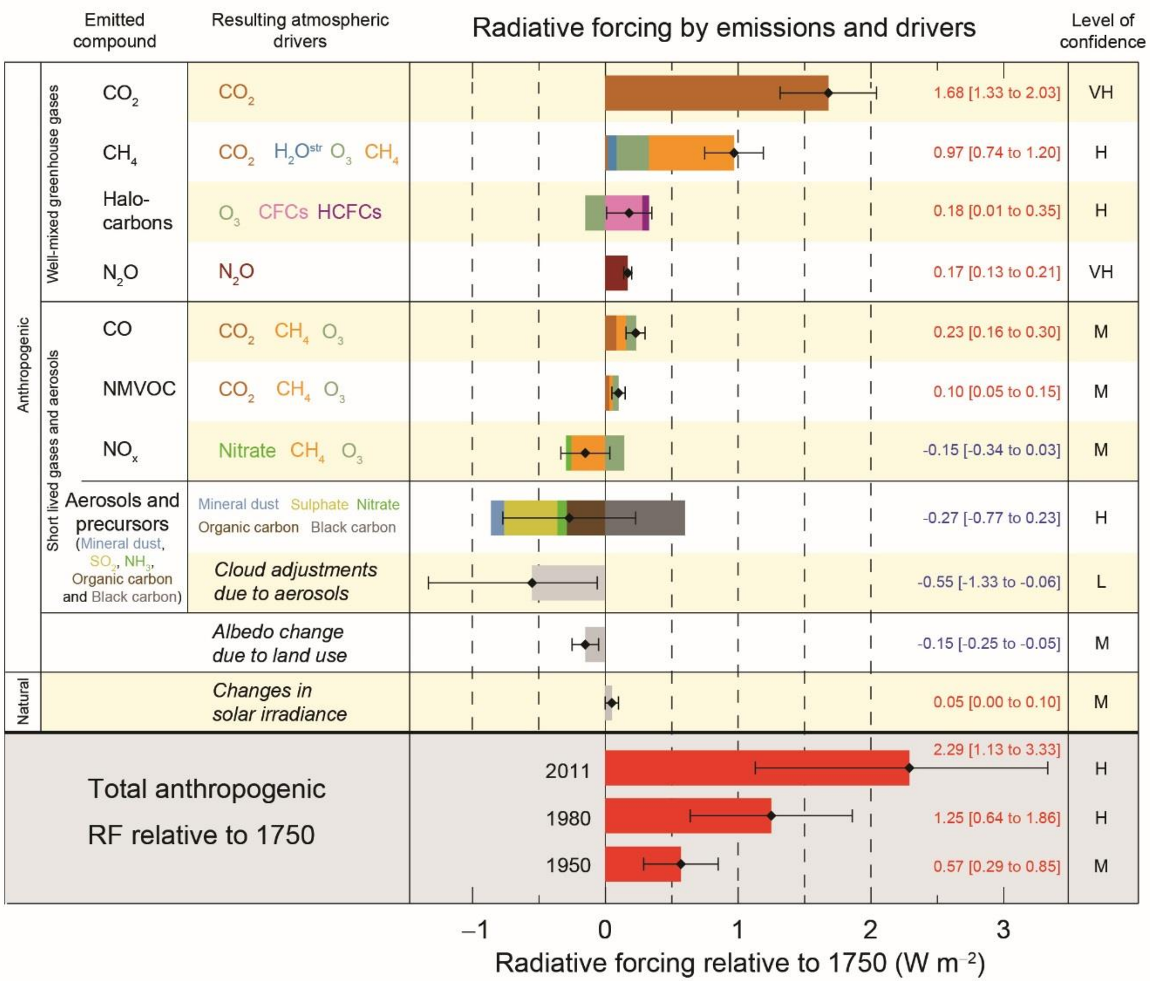
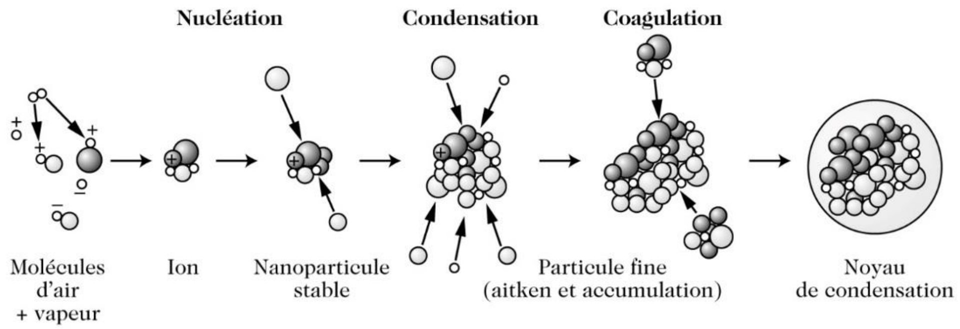
**Acknowledgments**
Co-workers, CNRS, Région Rhône-Alpes, FRAMA

# Motivation: air quality and climate




IPCC report 2013, http://www.climatechange2013.org/images/figures/WGI_AR5_FigSPM-5.jpg

# Motivation: fundamental processes




**How particles are formed in the atmosphere? Fundamental issues still remain !**

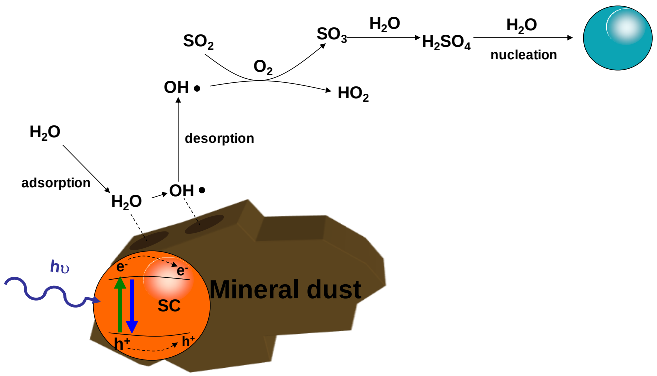
- **Nucleation takes place everywhere in the atmosphere !**
- **Nucleation involves $H_2SO_4$ and Ammonia nucleation in the gas phase**:

  Kirkby et al., Nature 2011, Kulmala et al., Nature 2012.

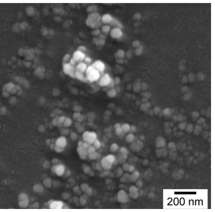
**Other pathway:**

- **Mineral dust at low concentration promotes $H_2SO_4$ nucleation in the gas phase :**

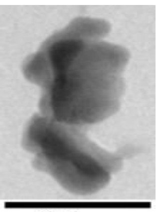
  Dupart et al., PNAS 2012.



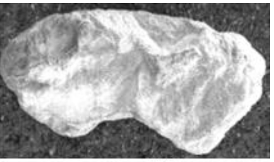
# Motivation: atmosphere mass content above PBL




Soot particle [ILM]




Dust particle [ILM]




Volacanic ash particle [Munoz]

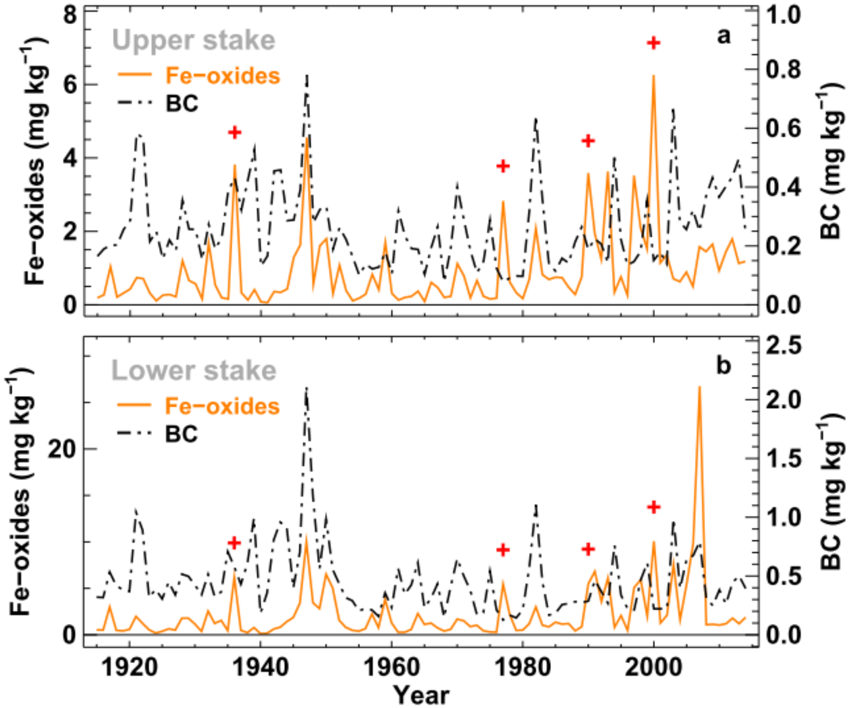



**Figure 4.** Average Fe-oxide and BC concentrations in the surface snow at the upper and lower measurement site on Claridenfirn for the period 1914–2014. The crosses mark years with exceptionally high Saharan dust activity. Note that the scales for upper and lower stake are different. [Gabbi et al. 2015]

**Les effets de la déposition des particules de suies et des poussières de sable** [ Gabbi et al., 2015, Jacobi et al. 2015]

- **Augmentation de 15-19 % du taux de fonte des glaciers**
- **Diminution de l'albédo des glaciers**
- **Forçage radiatif :**
    - **Suies: [-0,5; -1,5 w/m2]**
    - **Poussières de sable: [+1,5; 2 W/m2]**

# Outline

**CONTRIBUTION OF LASER REMOTE SENSING TO IMPROVE THE KNOWLEDGE ON THESE TOPICS ?**

1. CARBON AEROSOL

2. .1 Remote sensing the light interaction with the atmosphere

1.2 Laser induced incandescence (LII) on Light Absorbing Particles (LAP)

1.3 Coupling LII and Lidar

1.4 Field test measurement

2. PARTICULATE MATTER AND PARTICLES NUCLEATION

2.1 Particles nucleation observation with Lidar: sensitivity study

2.2 UV Polarization Lidar

2.3 Field test measurements

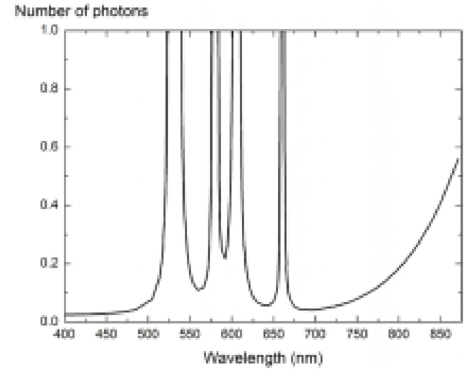
2.4 Particulate matter advection from local and regional sources

3. GREENHOUSE GASES: CH4, H20

3.1 OSAS a new methdoldoly

3.2 Validation experiment

3. CONCLUSION AND OUTLOOKS

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June 2014

Spotlight Summary by Katrianne Lehtipalo

**UV polarization lidar for remote sensing new particles formation in the atmosphere**

Atmospheric aerosol particles worsen the air quality and reduce atmospheric visibility. According to WHO, air pollution is the biggest environmental health risk. On the other hand, aerosol particles have a net cooling effect on the climate, thus counteracting the increase in greenhouse gases. Secondary particle formation from precursor vapors, often called 'new particle formation', is a major source of ultrafine particles (<100 nm) and cloud condensation nuclei. New particle formation seems to occur all around the world in many different kinds of environments, both in the planetary boundary layer and in the free troposphere. It involves a complicated chain of reactions and processes, which are not yet understood in detail.

Detecting new particle formation is technically challenging, mainly due to the small size of the newly formed particles and low concentrations of the relevant precursor vapors. A range of in-situ measurement techniques have been developed in recent years, but observing new particle formation with remote sensing technology is still work in progress. Thus observations of new particle formation in the free troposphere have been limited to sporadic measurements at mountain locations or airplane campaigns.

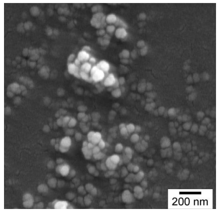
**Article Information**

UV polarization lidar for remote sensing new particles formation in the atmosphere

Grégory David, Benjamin Thomas, Yoan Dupart, Barbara D'Anna, Christian George, Alain Miffre, and Patrick Rairoux

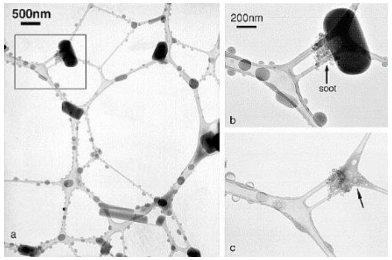
Opt. Express **22**(S3) A1009-A1022 (2014) View: Abstract | HTML | PDF

# Carbon aerosol

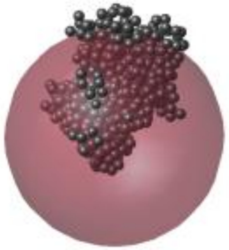



ILM, soot particles

**What is carbon aerosol?** soot particles, complex carbonaceous particles, biomass burning or secondary organic aerosols




Biomass burning particles
Jia Li et al. GRL, 2003



Light Absorbing Carbon
M. Kahnert, Opt. Expr. 2013

**LAC: Light-Absorbing Carbon particles, Black Carbon**.

T. Bond and R. Bergstrom, review in J. Aero. Sci. and Tech., 2006.

**LAP: Light absorbing particles**

Miffre et al. 2015.

**Optical Properties:**

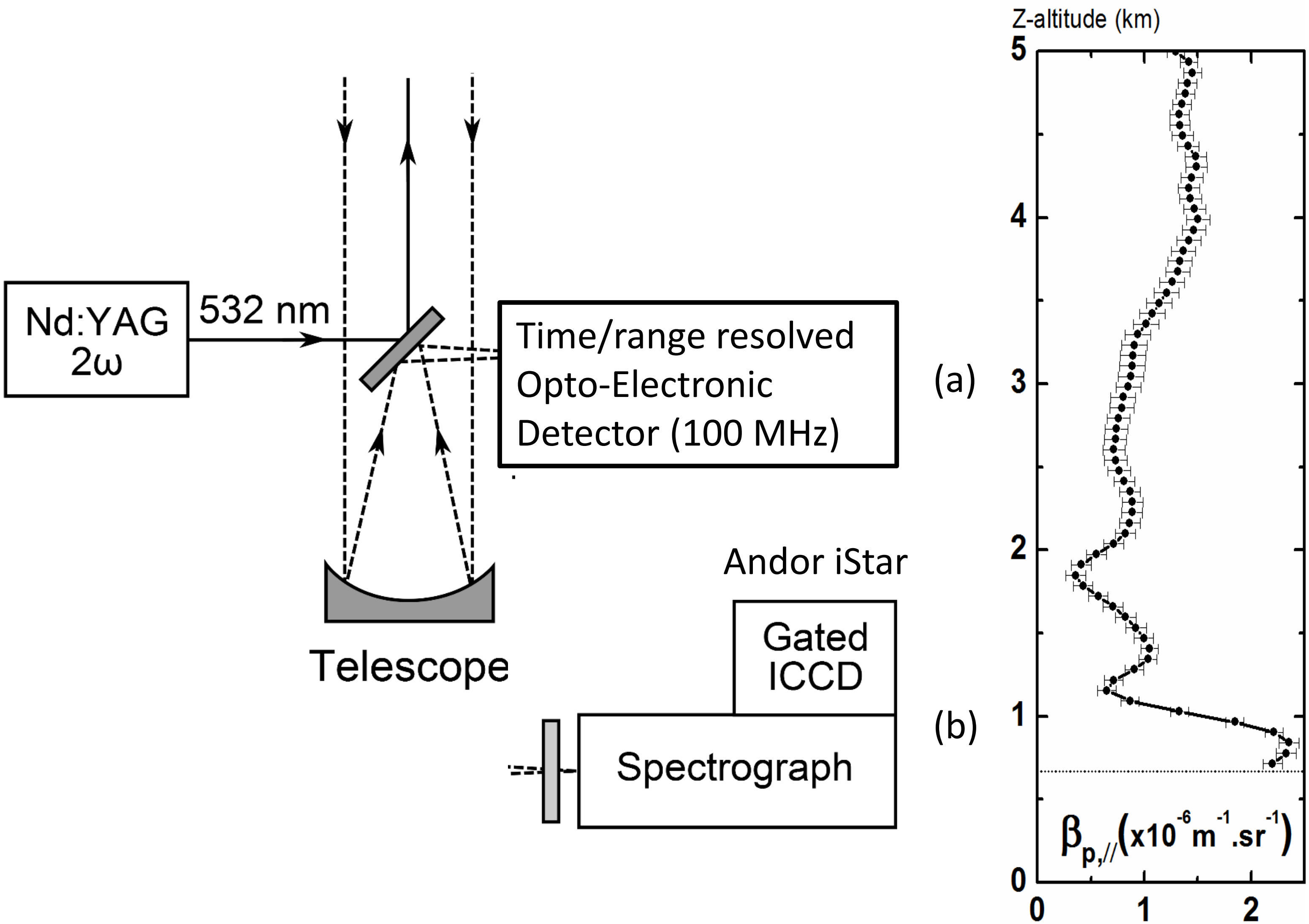
M.I. Mishchenko et al. JQSRT, 2011, M. Kahnert et al. Opt. Expr. 2013.

**Lidar Remote Sensing of biomass burning particles.**

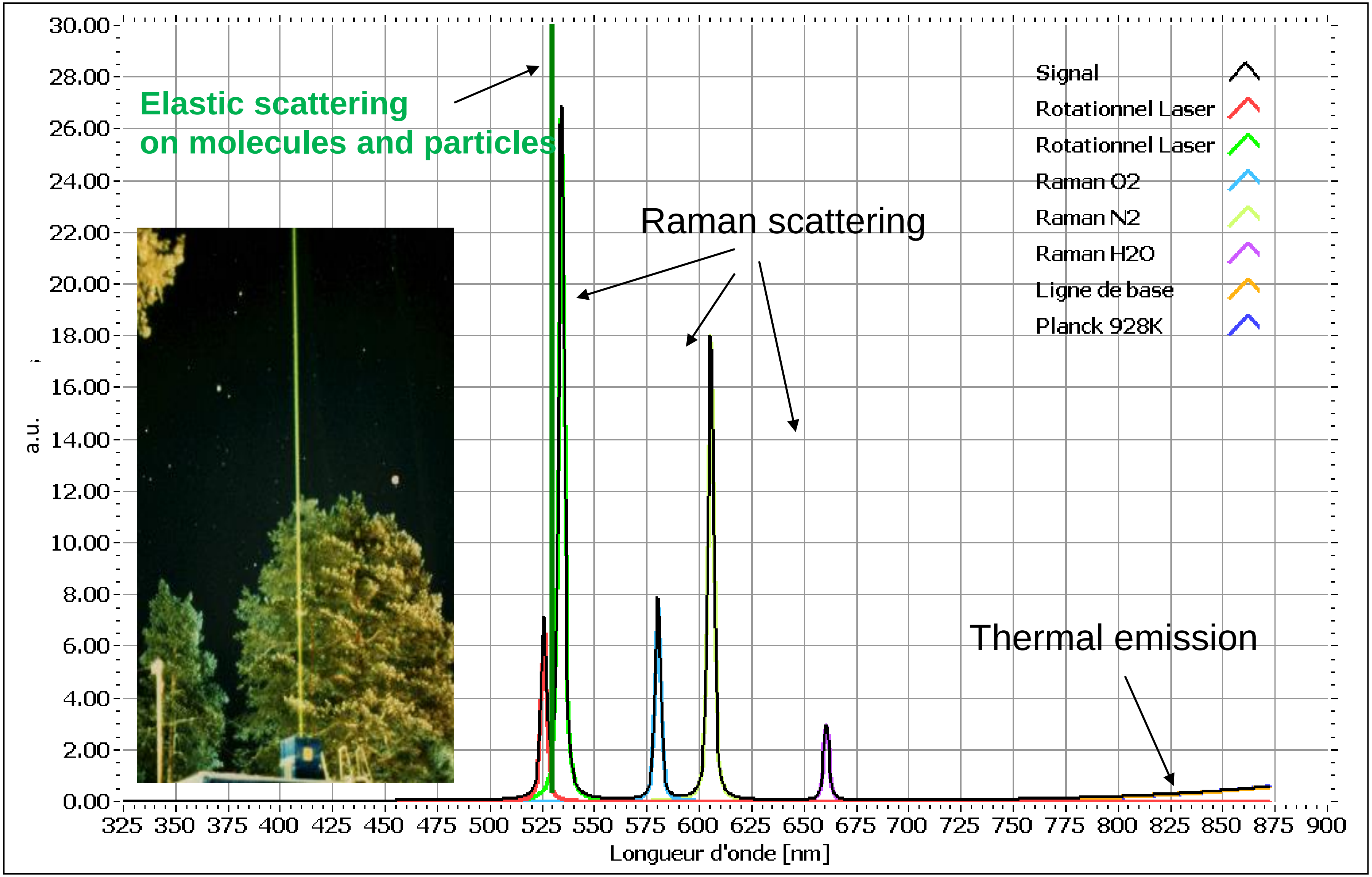
**Elastic and inelastic scattering.**

Igor Veselovskii et al. ACP 2015. “Characterization of forest fire smoke event near Washington, DC in summer 2013 with multi-wavelength lidar “

# Remote sensing the light interaction with the atmosphere

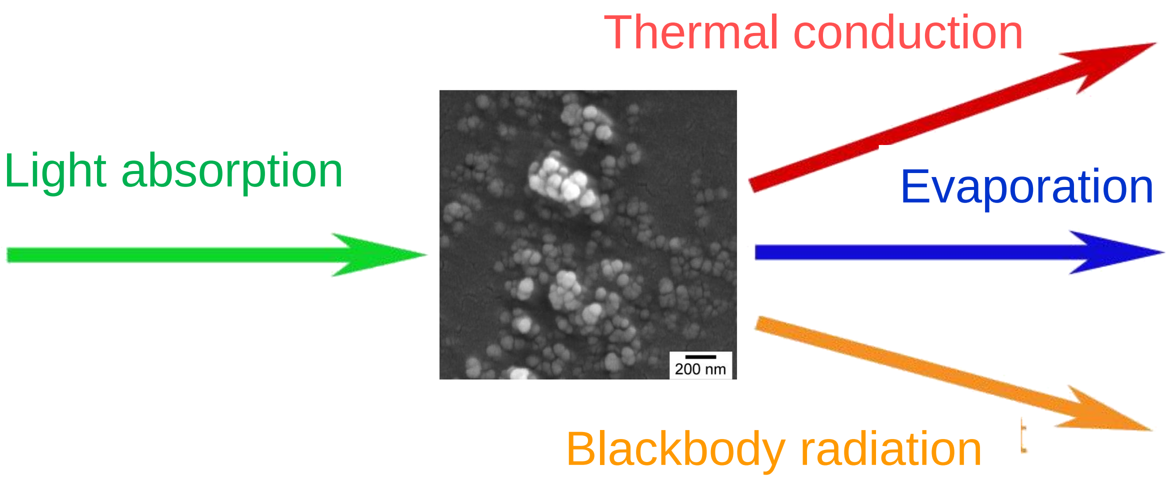



# Laser light interaction with the atmosphere: the spectrum




Atmosphere Spectrum  from an air parcel located at 300 m altitude above ground level at Lyon

# Laser Induced Incandescence (LII)

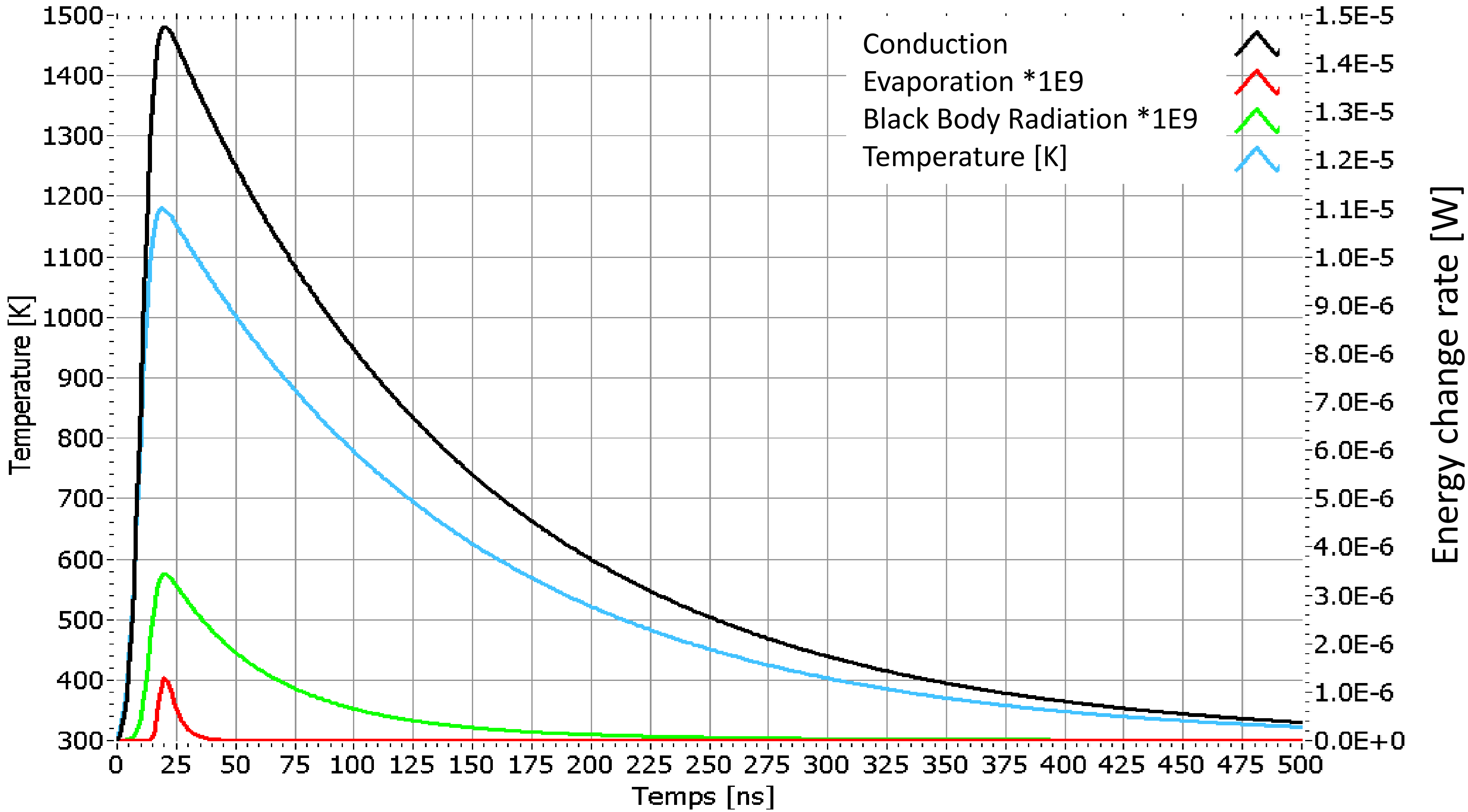



Absorption – Conduction – Evaporation – Blackbody radiation = Internal energy

$$Q_{abs}.\pi.a^2.q(t)-4.\pi.(T-T_0).\Lambda-\frac{\Delta H_v}{W_s}.\frac{dM}{dt}-4.\pi.a^2.\varepsilon.\sigma.(T^4-T_0^4)=\frac{4}{3}.\pi.a^3.\rho_s.C_s.\frac{dT}{dt}$$

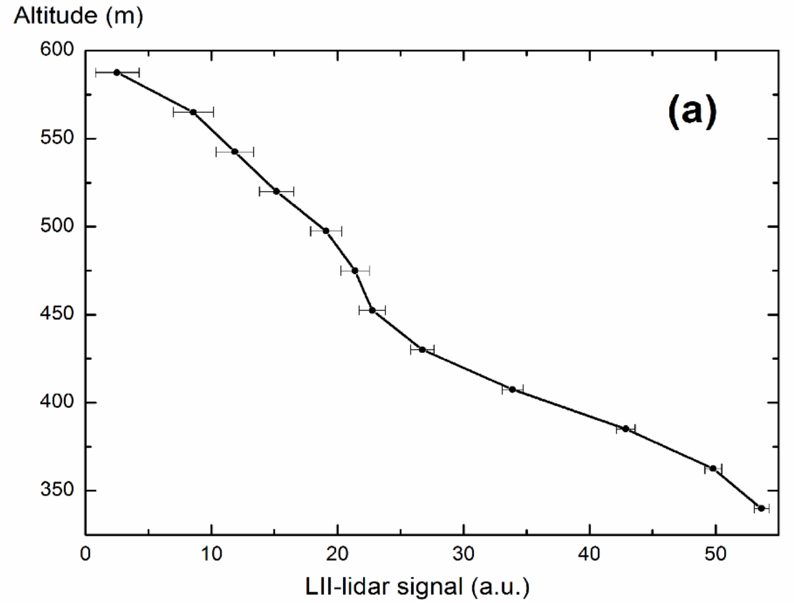
C. Schultz et al., Special issue, Appl. Phys., B (2006). G. Zizak et al., Special issue, Appl. Phys., (2011).

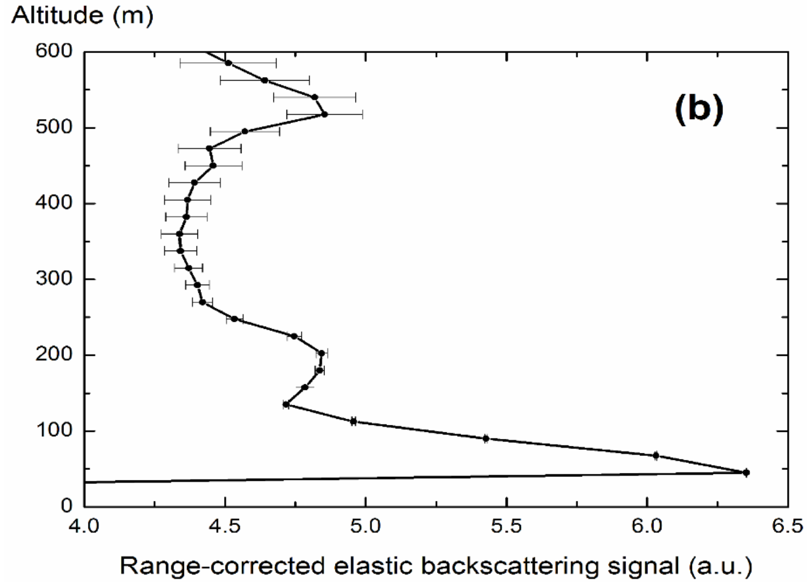
# LII Simulation for a single Light-Absorbing Particle (LAP)



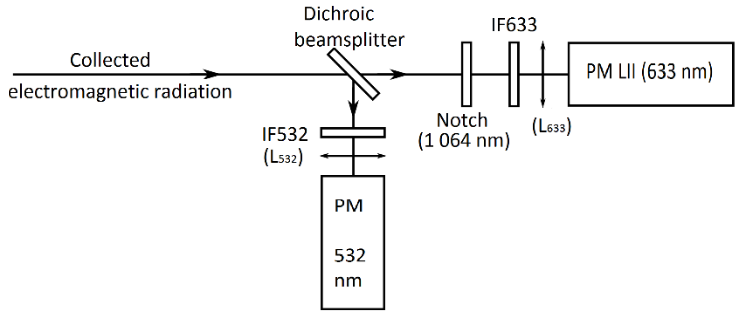

Particle diameter 50 nm, laser excitation at 532 nm, pulse duration = 20 ns, I = 2,8.10$^{8}$ W.cm$^{-2}$

# LII Lidar experiment : Soot content in the atmosphere




(a) Remote sensing of black body emission from the **soot particles** heated with a pulse laser: **LII Signal** proportional to the **Soot Particles mass concentration.**




**(b) Elastic backscattering-lidar signal from the total particles content**

# Aerosols load monitoring in urban area with UV-Lidar and air masses trajectories (1)

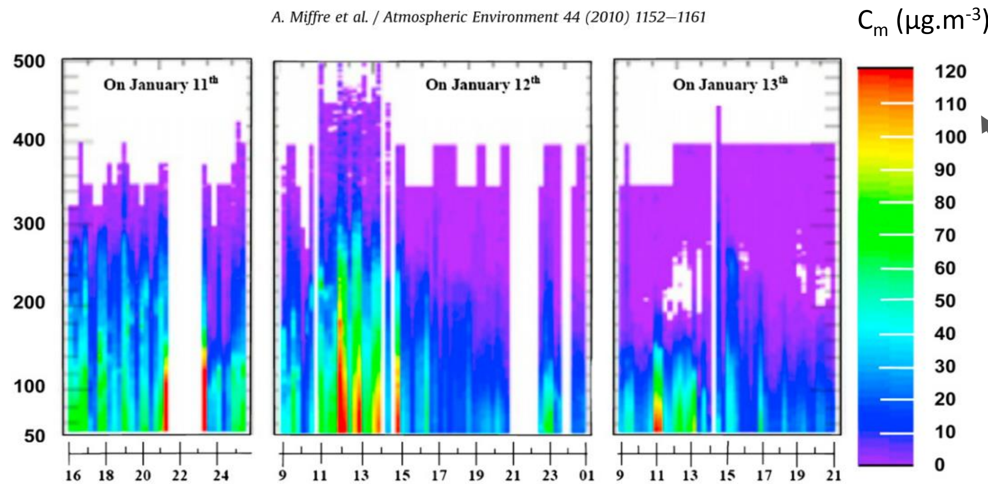
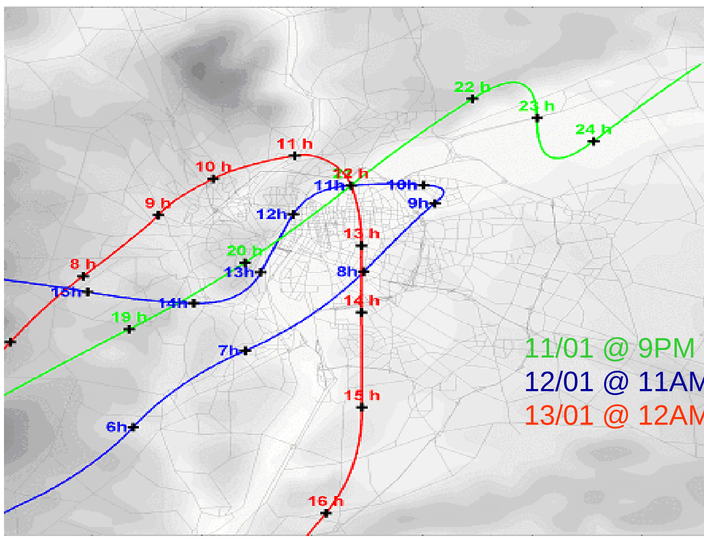
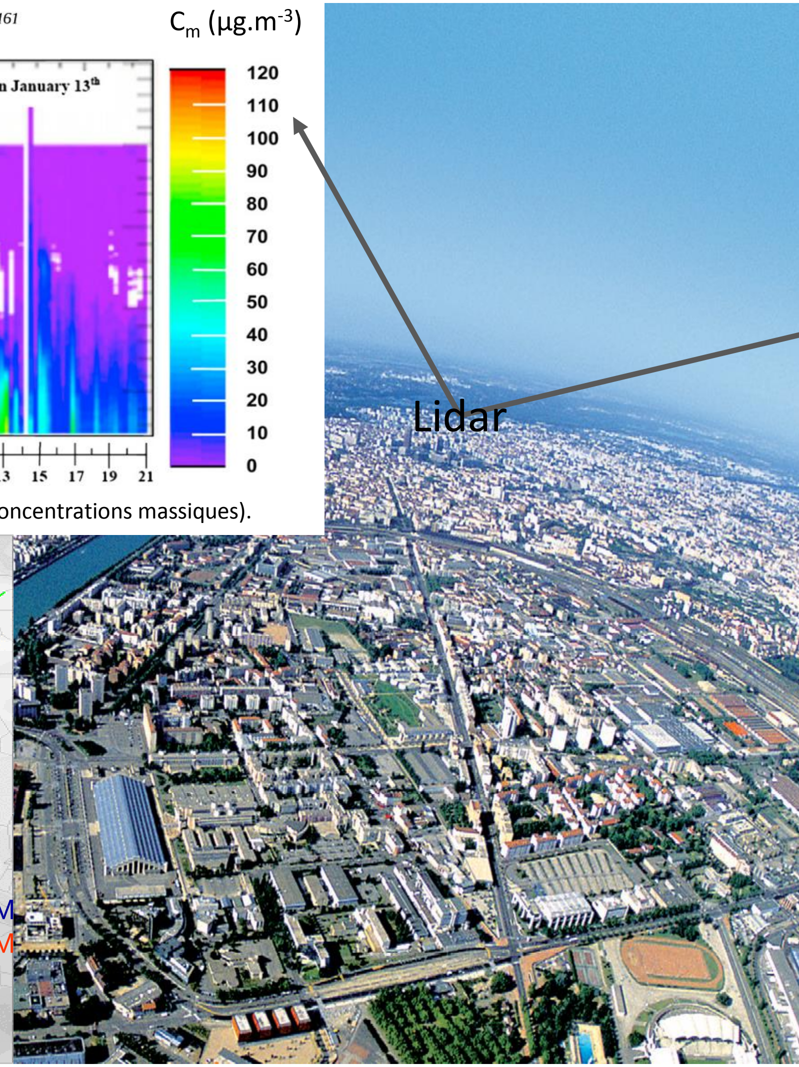



Fig. 5: Cartographie temps-altitude de la charge en aérosols urbains (concentrations massiques).

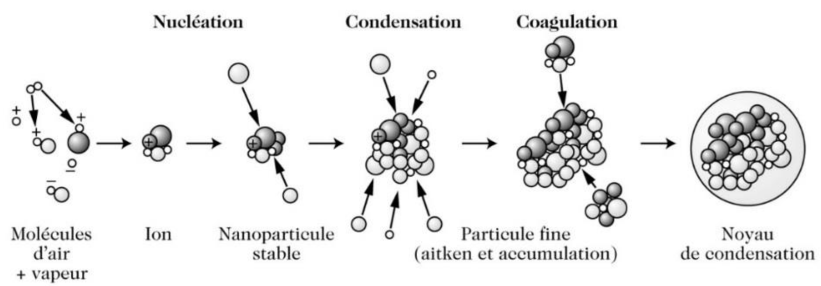



Photographie aérienne de la ville de Lyon.

[1] A. Miffre, M. Abou Chacra, S. Geffroy, L. Soulhac, R. Perkins, E. Frejafon and P. Rairoux, *Atm. Env.*, (2010).

# 2. Nucleation in the atmosphere

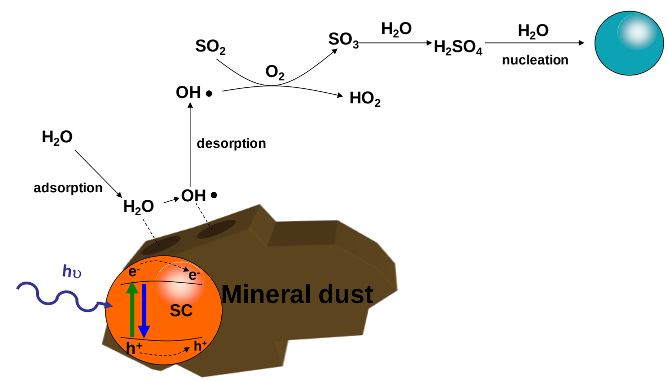



## Particles nucleation observation

**In the atmosphere**: Kulmala et al., (2012), Wiendesohler (2009)
Hamburger ACP 2011, Weigel, ACP (2011).

**In laboratory**: Kirkby et al., Nature 2011, Y. Dupart et al. PNAS 2012.

**Lidar observation**: K. Sassen, Env. Res. Lett. 2008



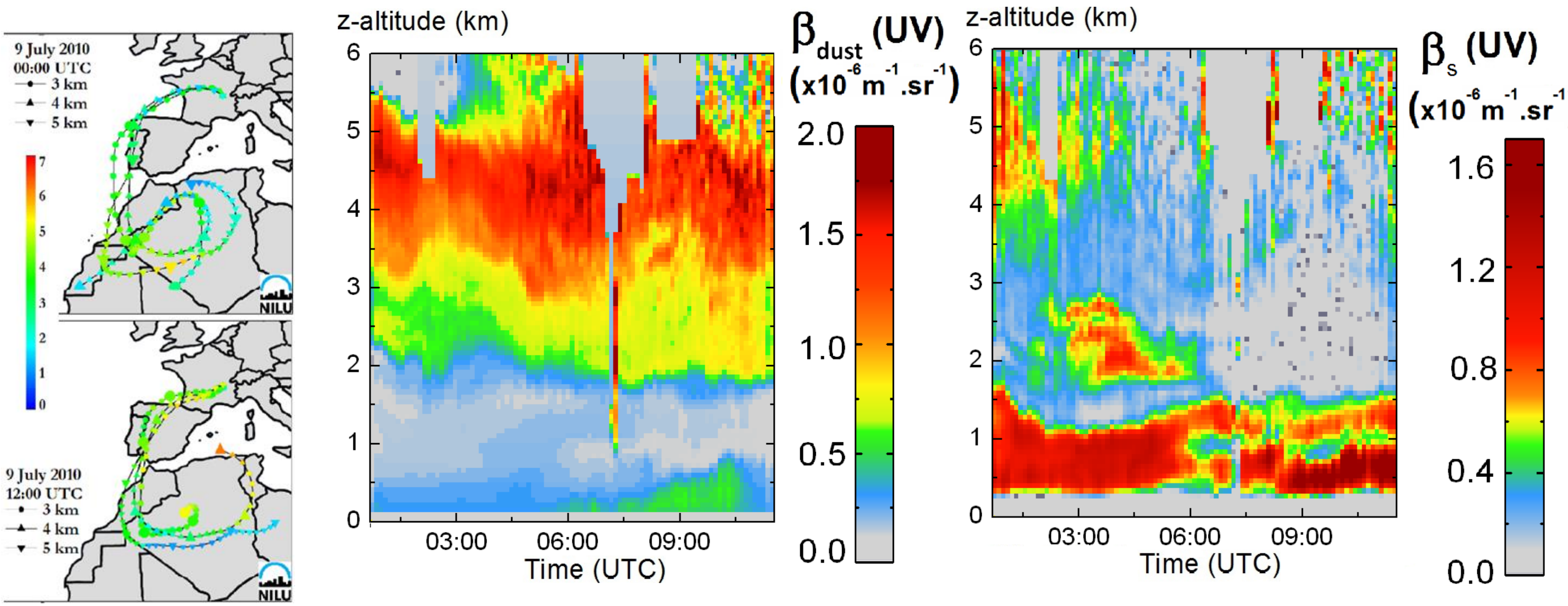


IOP Publishing — Environmental Research Letters
Environ. Res. Lett. 3 (2008) 025006 (12pp) — doi:10.1088/1748-9326/3/2/025006


### Cloud effects from boreal forest fire smoke: evidence for ice nucleation from polarization lidar data and cloud model simulations

Kenneth Sassen[1] and Vitaly I Khvorostyanov[2]

# UV-polarization backscattering during desert dust outbreak [1]

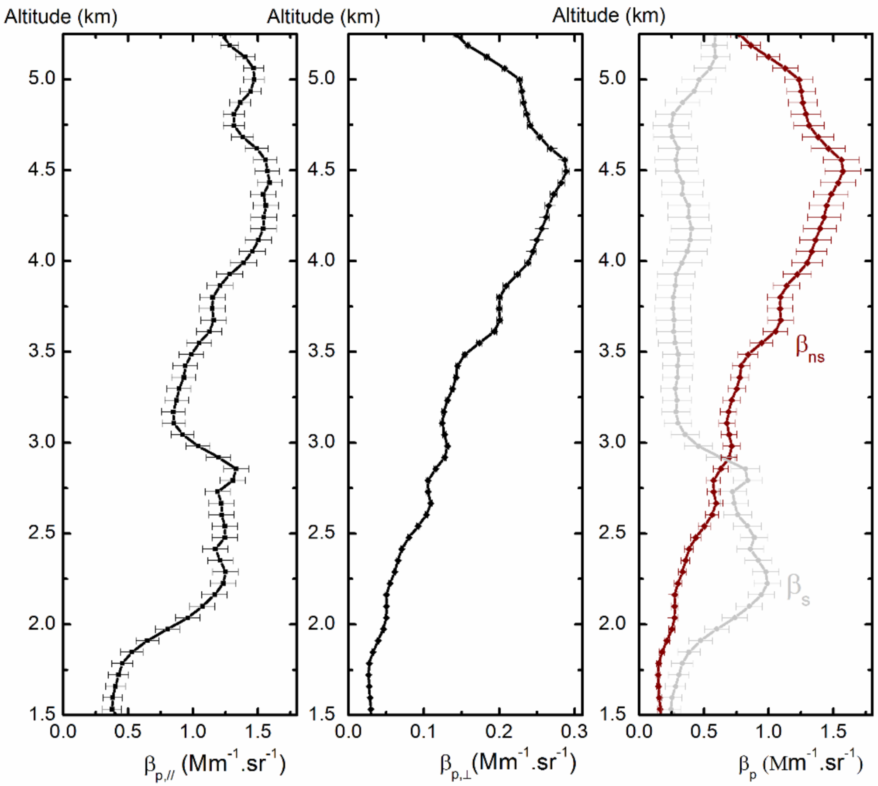


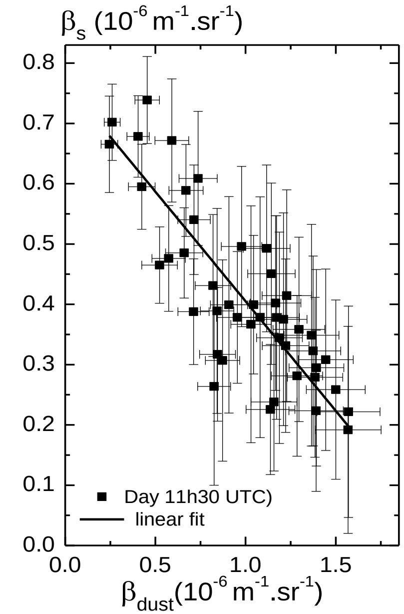

Non-spherical (dust) and spherical (s) particles backscattering in the atmosphere, Lyon 2012,
From the UV-pol measurement by applying the two-components partitioning formalism[2].

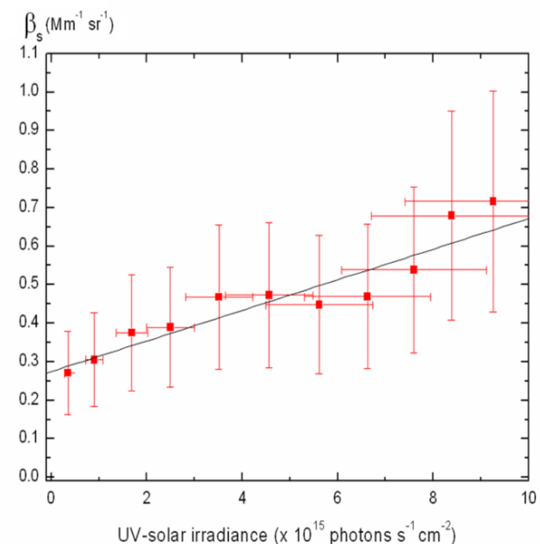
iLM Institut Lumière Matière

[1]G. David, A. Miffre et al. OE, 2014, [2]A. Miffre et al., GRL 2011.

# UV-polarization backscattering:  NPF event analysis



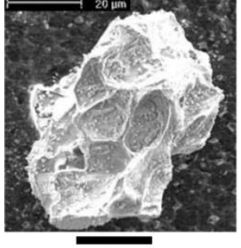

- Particles backscattering partitioning allows observing the NPF event [2]. $\beta_{dust} = \beta_{ns}$

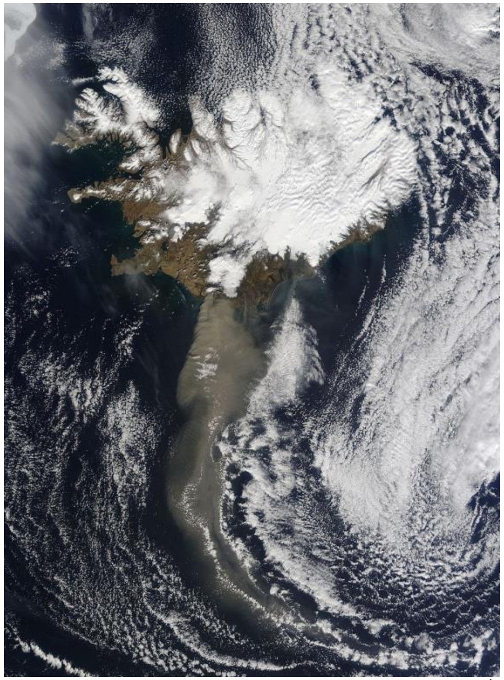
- Assumption, above 2 km: $\beta_{NPF} = \beta_S$

-Field measurements exhibit the same features as Laboratory experiments on new particle formation (NPF) C. George's group, [1] and H. Hermann ground based measurements[1]

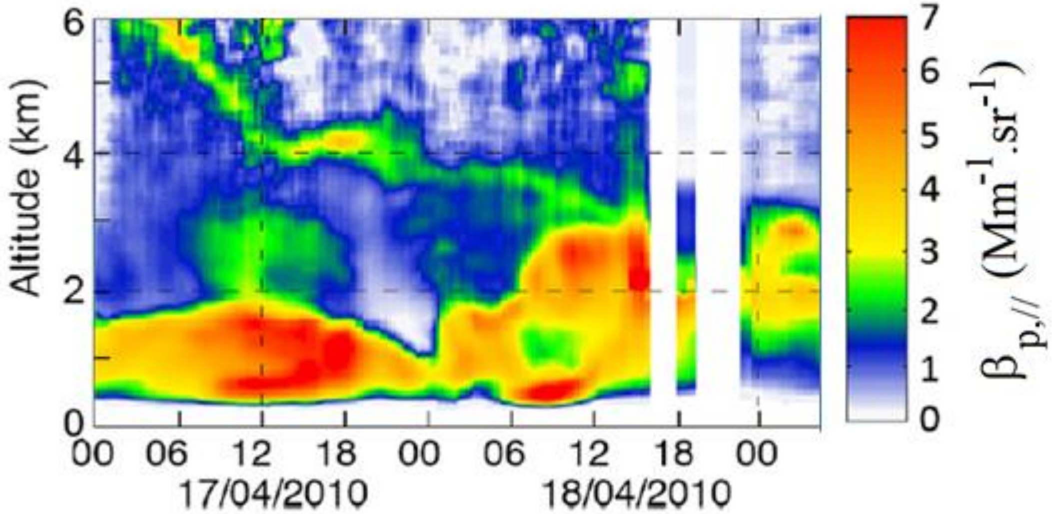
[1] Y. Dupart et al., *PNAS* (2012), [2] David et al. Opt. Expr. 2014

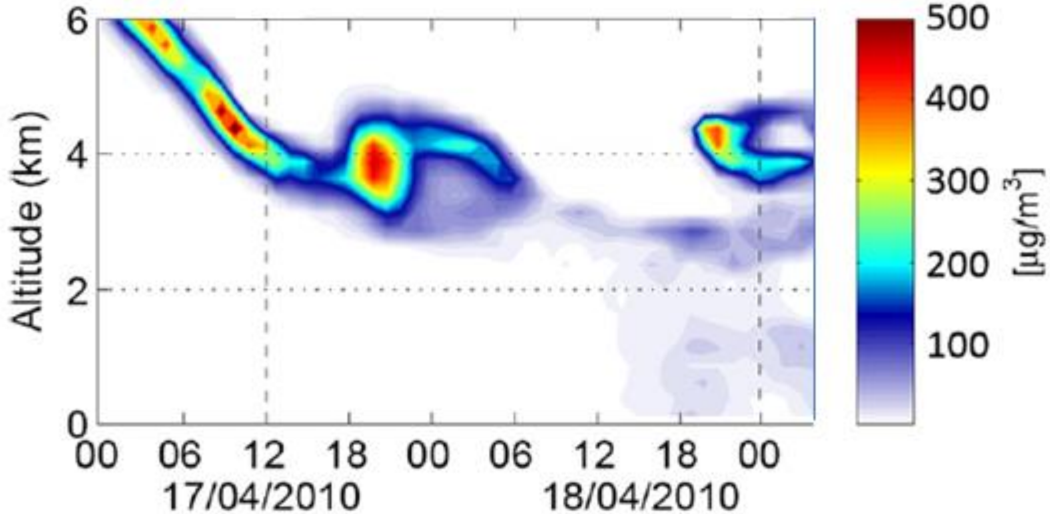
# Volcanic ash monitoring above Lyon during the Eyjafjallajökull volcano eruption in April 2010



Eyjafjallajökull volcano eruption on April 19th 2010
(NASA, Earth observatory)

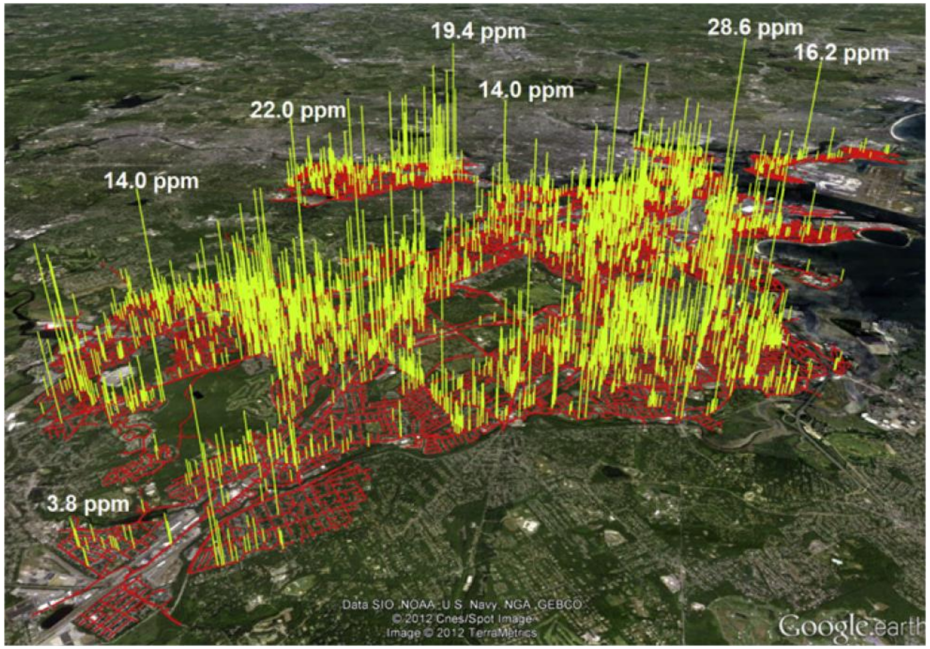


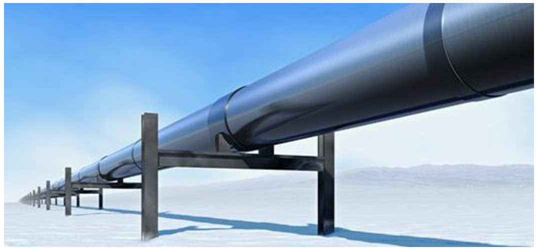
# 3. Laser remote sensing of greenhouse gases: CH4

**Naturelles : 180 - 270 Tg/yr**

**Anthropiques : 280 - 410 Tg/yr**




Phillips et al., *Environmental Pollution* (2013)



# Optical Similitude Absorption Spectroscopy (OSAS): a new methodology

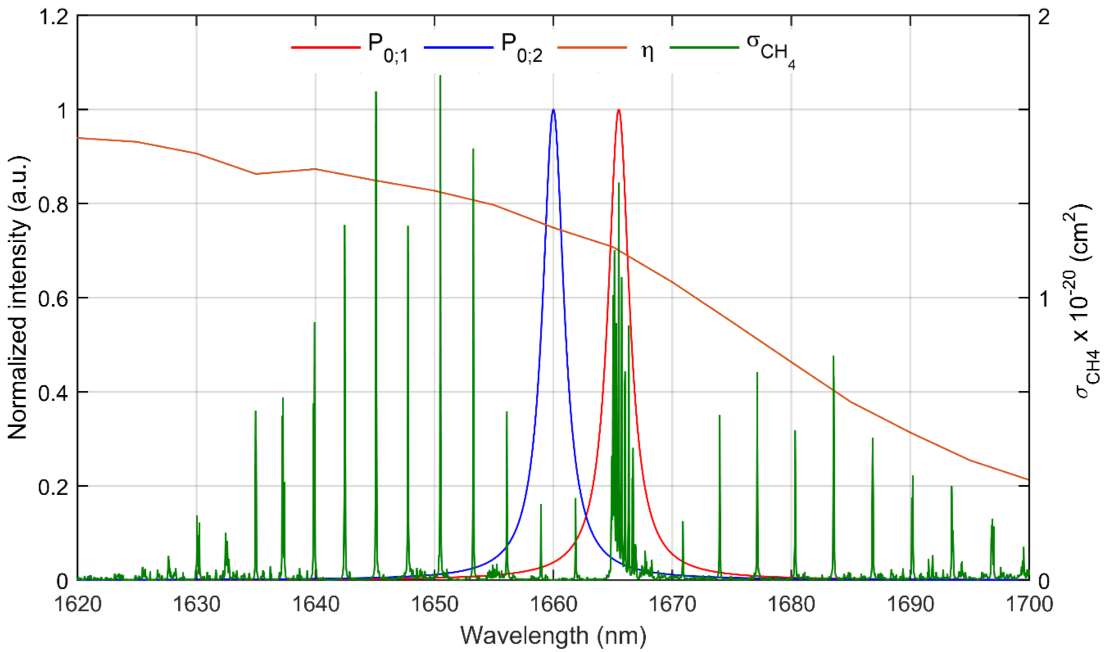
## Méthodologie OSAS

*Mesure d'absorption différentielle à faible résolution spectrale*

$P_{0;1}(\lambda)$ : actif/similaire
$P_{0;2}(\lambda)$ : reference

$T(\lambda)$ $\eta(\lambda)$ $S_1, S_2$

Source(s) De lumière → Volume de mesure → Photo-détecteur → Système d'acquisition




$$S_i = K \int_{\Delta\lambda} P_{0;i}(\lambda) T(\lambda) \eta(\lambda)\, \mathrm{d}\lambda$$

i = 1, 2

$$T(\lambda) = \exp\left[-N\sigma(\lambda)\ell\right]$$

$K$ : Constante opto-électronique

$P_{0;i}(\lambda)$ : Densité spectrale

$N$ : Concentration volumique ($\#.cm^{-3}$)

$\sigma(\lambda)$ : Section efficace d'absorption ($cm^2$)

$\ell$ : Longueur du chemin optique(cm)

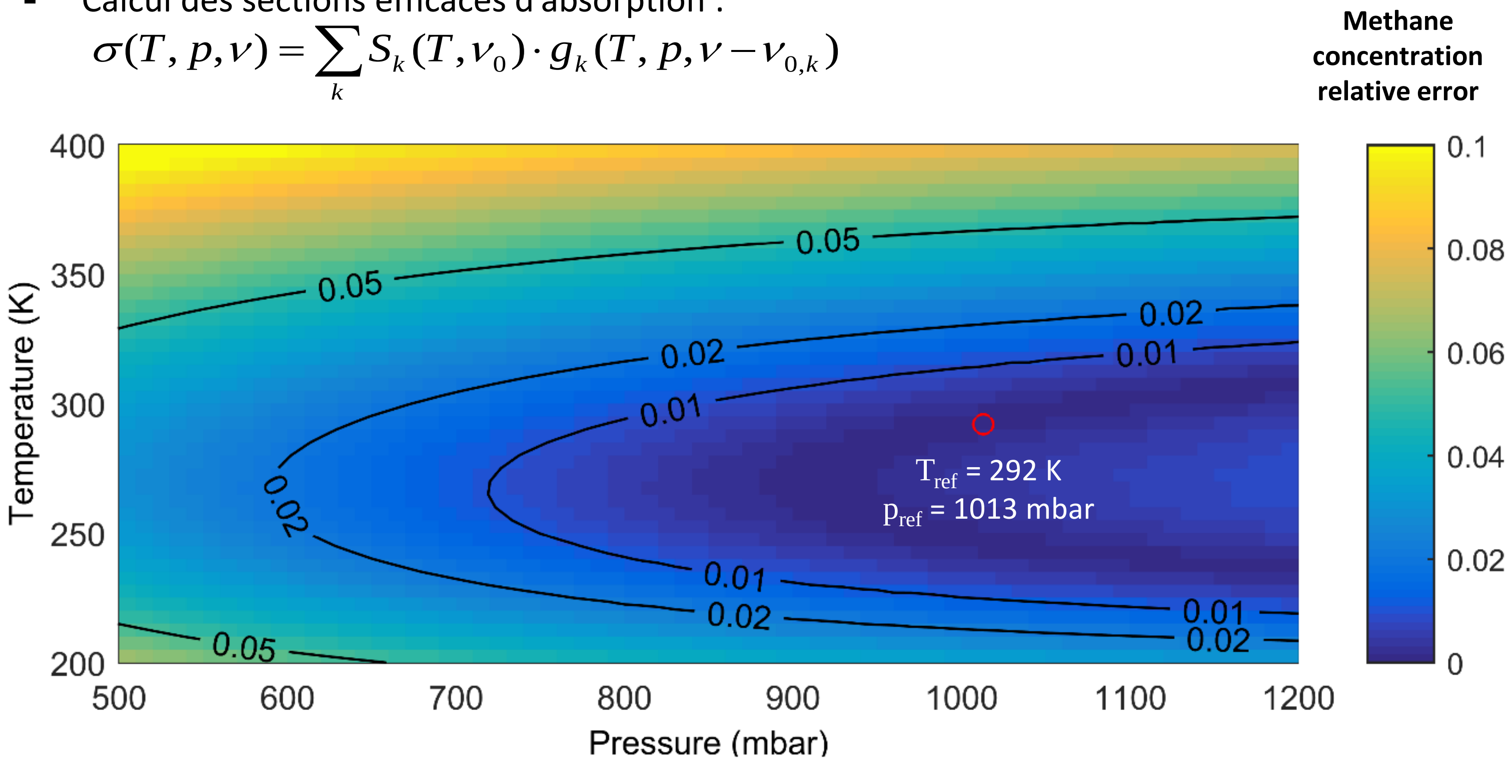
$\eta(\lambda)$ : Efficacité de détection

# Optical Similitude Absorption Spectroscopy (OSAS)

## Erreurs systématiques: σ(p , T)

- La température et la pression de l'atmosphère peuvent varier fortement, **cela n'affecte quasi pas la mesure**
  - Calcul des sections efficaces d'absorption :

$$\sigma(T, p, \nu) = \sum_k S_k(T, \nu_0) \cdot g_k(T, p, \nu - \nu_{0,k})$$

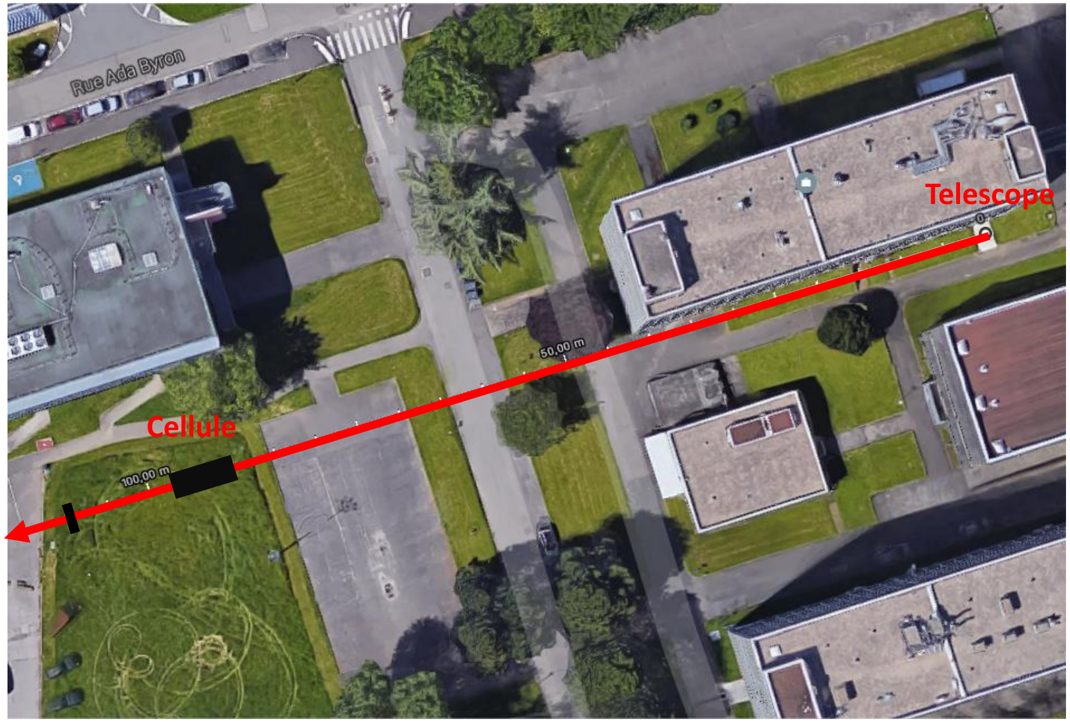



**Interprétation:**

- Effet de $\Delta p$: relativement faible (intégration du profil de raie)
- Effet de $\Delta$T: plus marqué (dépendance des forces de raies avec la température)

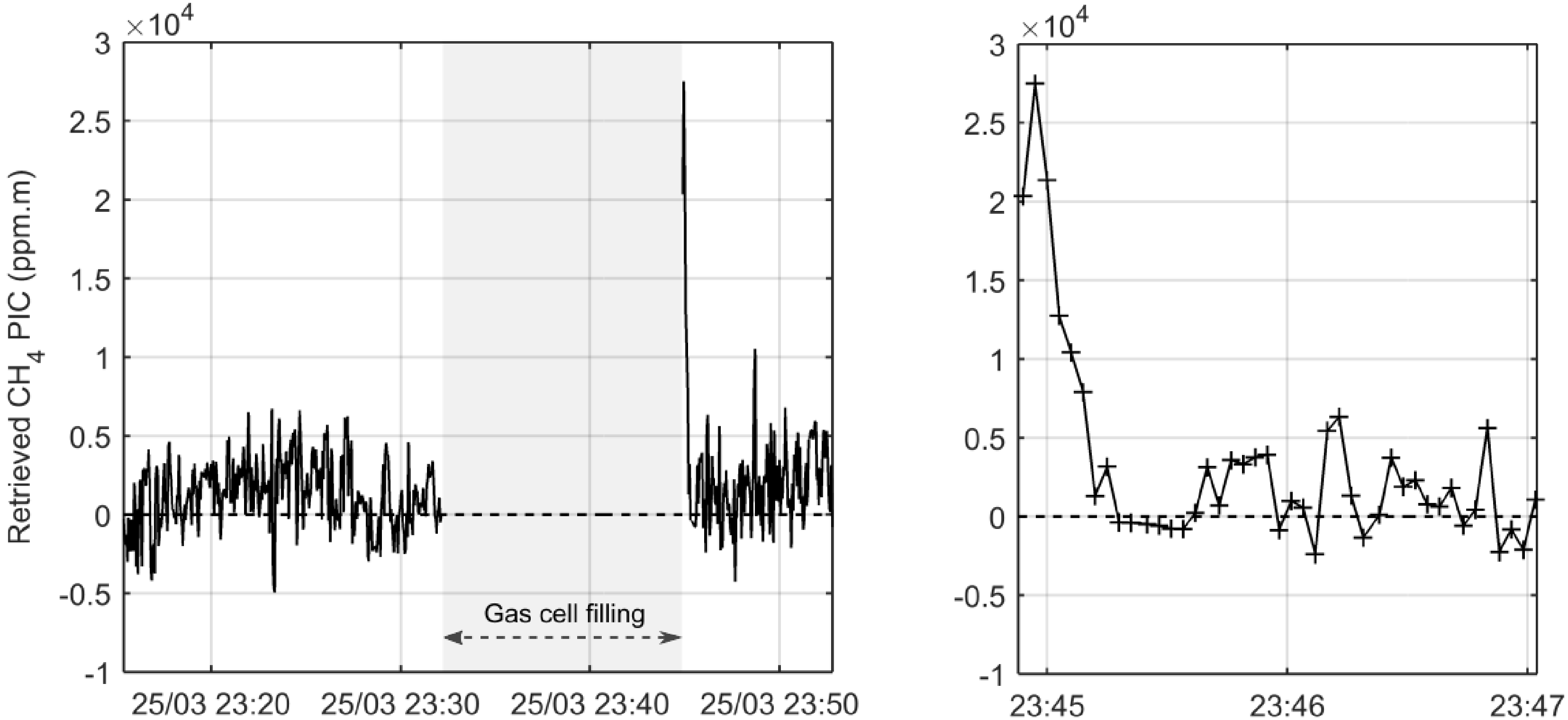
# Mesures en atmosphère extérieure

Réalisation d'une mesure de fuite de Méthane: validation




- Caractéristiques cellule: $\ell$ = 6 m, Ø = 60 cm
- Bouteille $CH_4$ = 1,8 %
- Ajout d'un réflecteur à $z$ = 110 m

# Résultats expérimentaux de validation




- Mesure rapide (1 secondes), dispersion du $CH_4$ en 20 secondes
- Cohérent avec la valeur maximale théorique de 10,8 × $10^4$ ppm.m (1,8 % × 6 m)
- Cohérente avec les mesures ponctuels
- Limite de détection: 2,2 × $10^3$ ppm.m (*en laboratoire: 0,65 ×* $10^3$ *ppm.m*)
- Amélioration: optique de détection (pas le but du travail présenté)

# Conclusion and outlooks

● Soot particles (LAP): Lidar remote sensing of Light Absorbing Particles (LAP) by detecting LII-thermal emission

- Validation with co-located instruments needed.

● NPF: Optical requirements to remotely observe with Lidar NPF-events promoted by non spherical desert dust particles/volcanic ash particles.

- single-scattering simulations/measurements.

● Long range and locac Advection: Lidar remote sensing of particulate matter transport into the atmosphere.

- Local, regional scale facilities
- Coupling with transport model gives information on the long range sources.
- Necessity of accurate and sensitive to ns-particles measurements.
- Scattering cross section from labs.

● Greenhouse gases.

- OSAS Lidar: $CH_4$ lake and diffuses sources detection. Experiment in the field !!
- Other traces detection: COVs, …

● Basic science and demonstration of future applications: it needs cooperation !! Welcome

EDUCATION: Master program UCBL-ECL; Sciences of Ocean Atmosphere and Climate (SOAC)

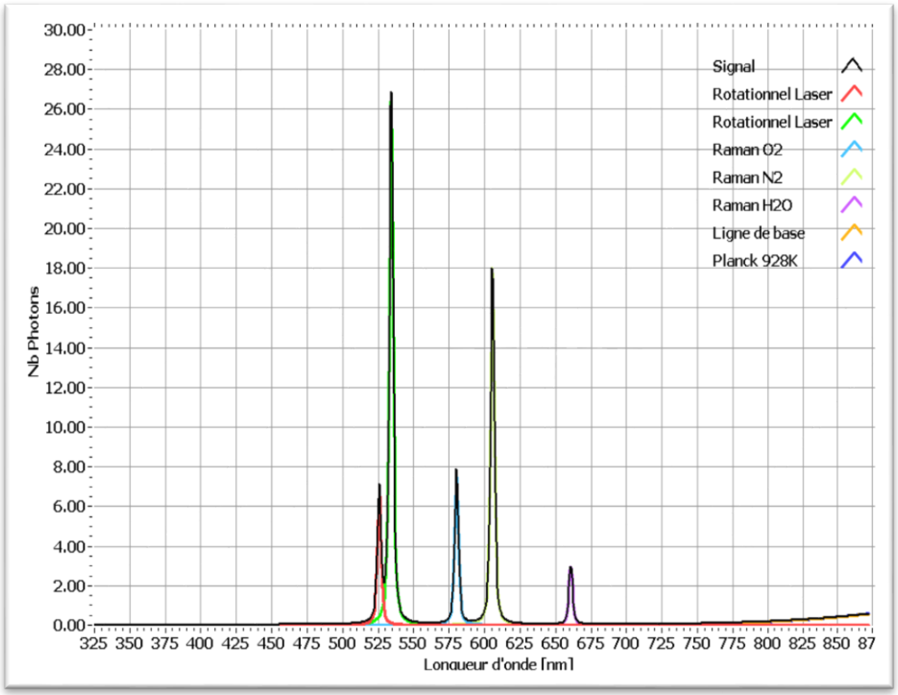
Thank you for your attention !

[1]G. David et al.,, Optic express, 2013. Miffre et al. JQSRT, 2015.

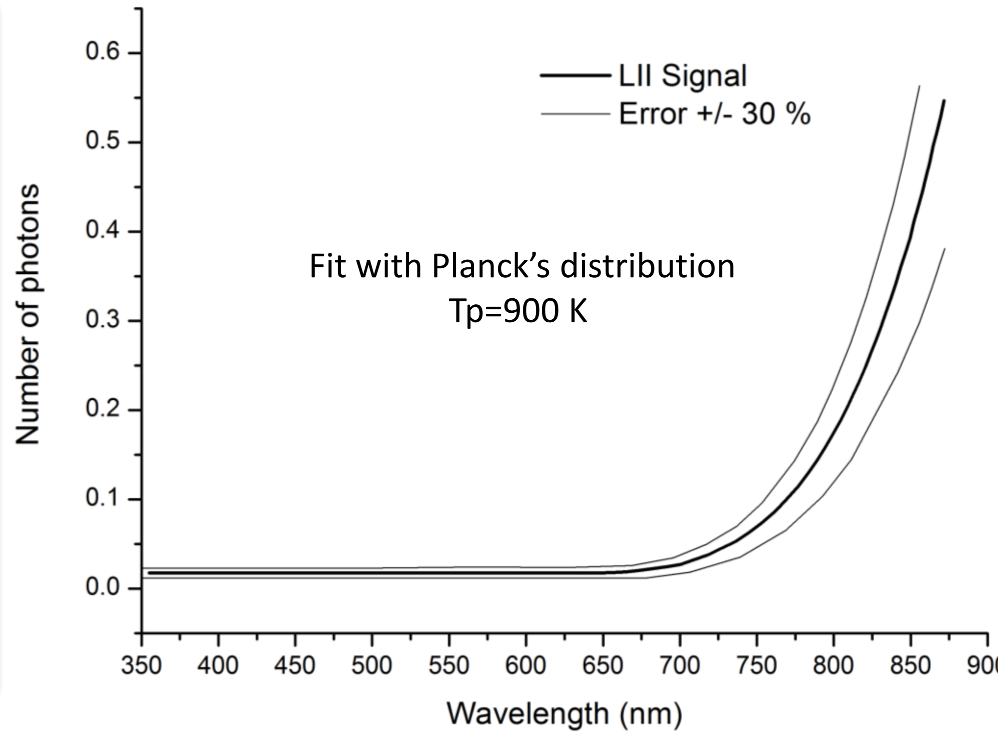
# SUPPLEMENTARY MATERIAL

# Lidar measurement of atmospheric particles thermal emission

Laser excitation at 532 nm




Laser excitation at 1064 nm




Lidar atmosphere spectrum: pulse duration: 20 ns, altitude range: 50-300 m
30 min acquisition time, Tp=928 K



A. Miffre et al. Opt. Expr (2015)

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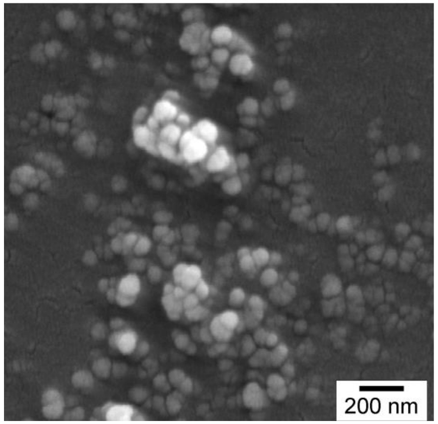
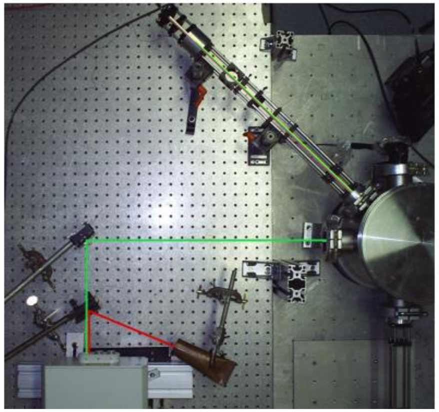
# LII measurement on LAP in laboratory (combustion soot)

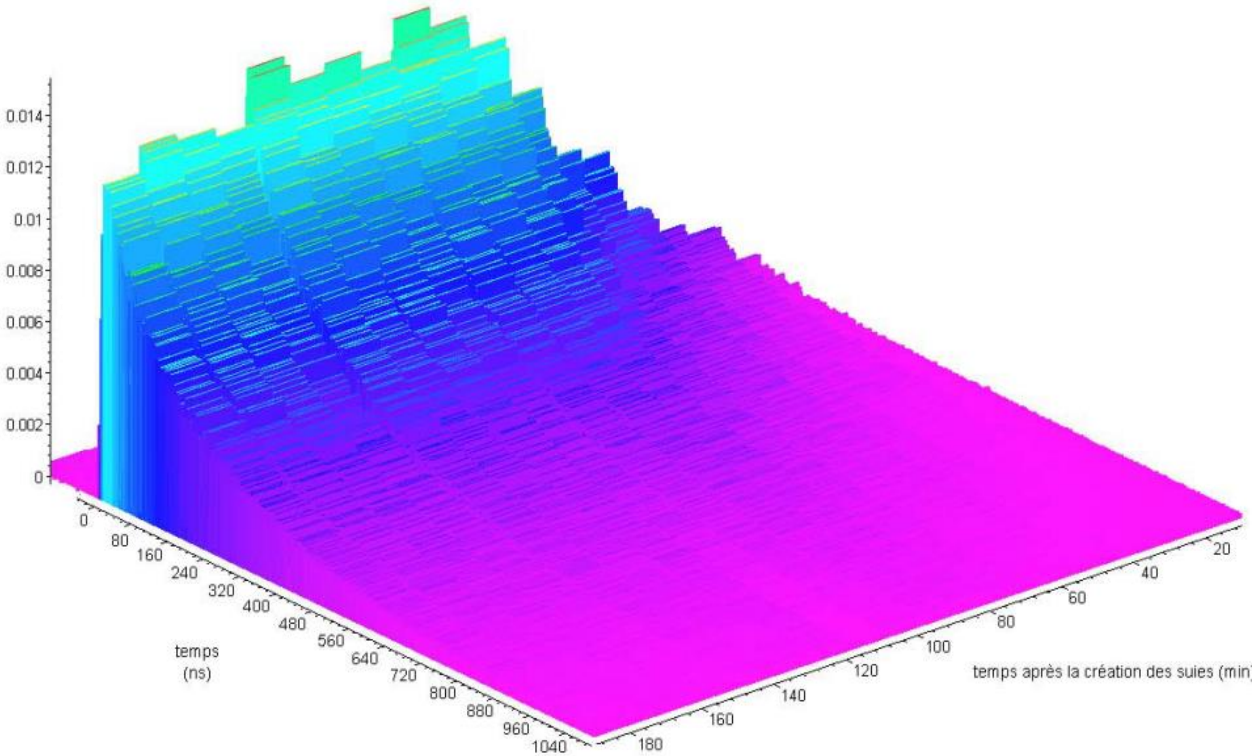
LII: Laser Induced Incandescence
LAP: Light-absorbing Particles




Soot Particles (ILM)



Laboratory experiment:
Laser excitation at 532 nm.
LII detection at 650 nm.




Measurement of the time dependence of LII signals. Signal detected at 650 nm.
LAP emission decay in the range of 200 ns. Experiment duration: 3 hours.

# Formalism for LII-Lidar remote sensing of LAP in the atmosphere

**LII for a single particle of radius *a*, Black Body radiation**

$$I_{LII}(\lambda,a) = \varepsilon_\lambda\, B_\lambda(T_p) \times 4\pi a^2$$

**Kirchhoff's law**

$$\sigma_{abs} = Q_{abs} \times \pi a^2 = \varepsilon_\lambda \times \pi a^2$$

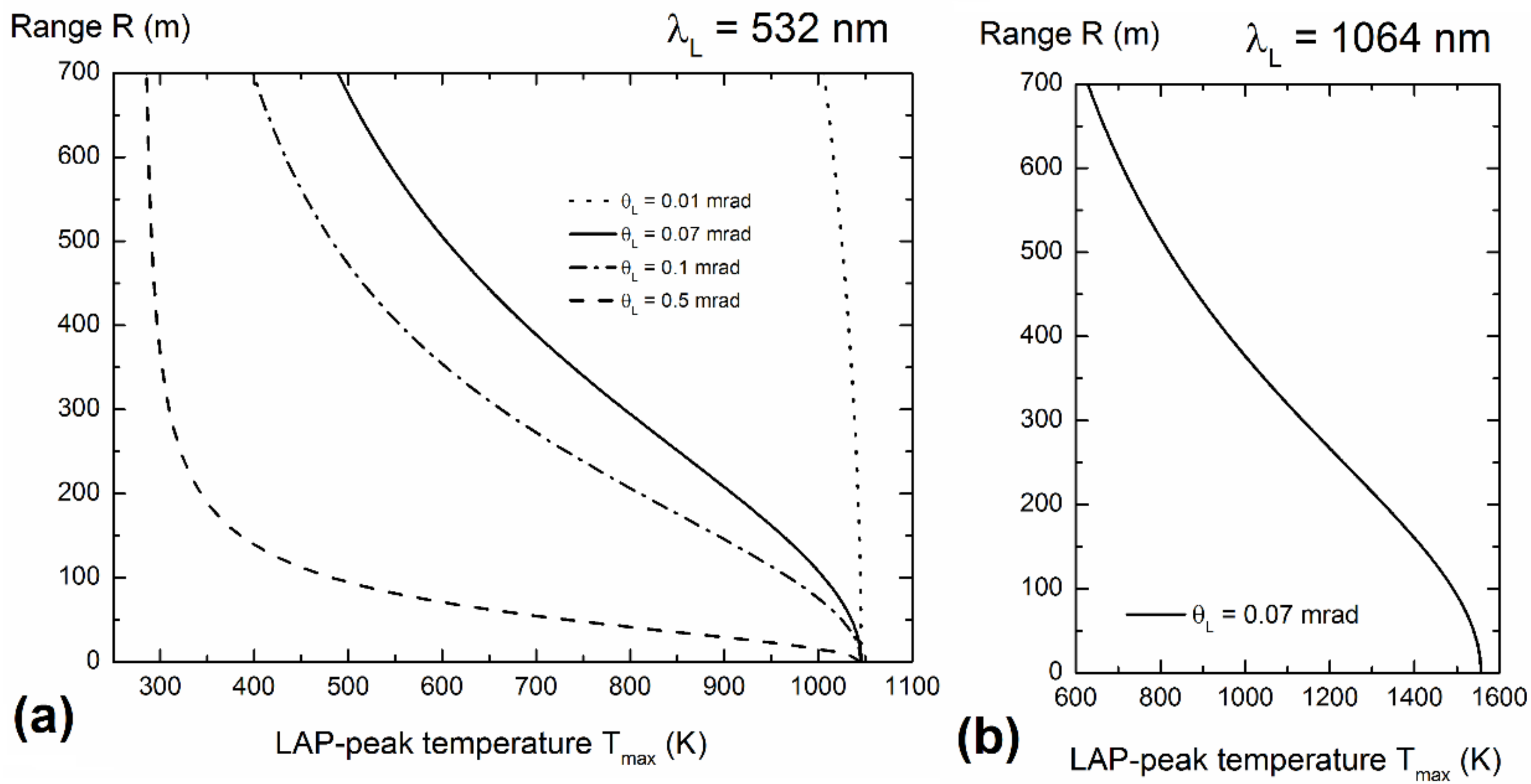
**LII reaching the lidar detector for a single particle of radius a located at the rang R.**

$$I_{LII}(\lambda,a,R) = I_{LII}(\lambda,a) \times \frac{A_0}{4\pi R^2} = 4\sigma_{abs} B_\lambda(T_p) \times \frac{A_0}{4\pi R^2}$$

**LAP max. Temperature**

$$T_{\max}(R) = T_a + \frac{6\pi E(m)}{\lambda_L \rho c_p} \frac{E_L T(\lambda_L,R)}{\pi(R^2\theta_L{}^2 + d_0^2)} \quad ; \quad E(m) = \mathrm{Im}((m^2-1)/(m^2+2))$$

a: particle size
$B_\lambda(T_p)$: Planck blackbody spectral radiance,
$E_l$: Laser energy,
T: atmosphere transmission
Tp: LAP temperature.
R: range
θ: laser divergence



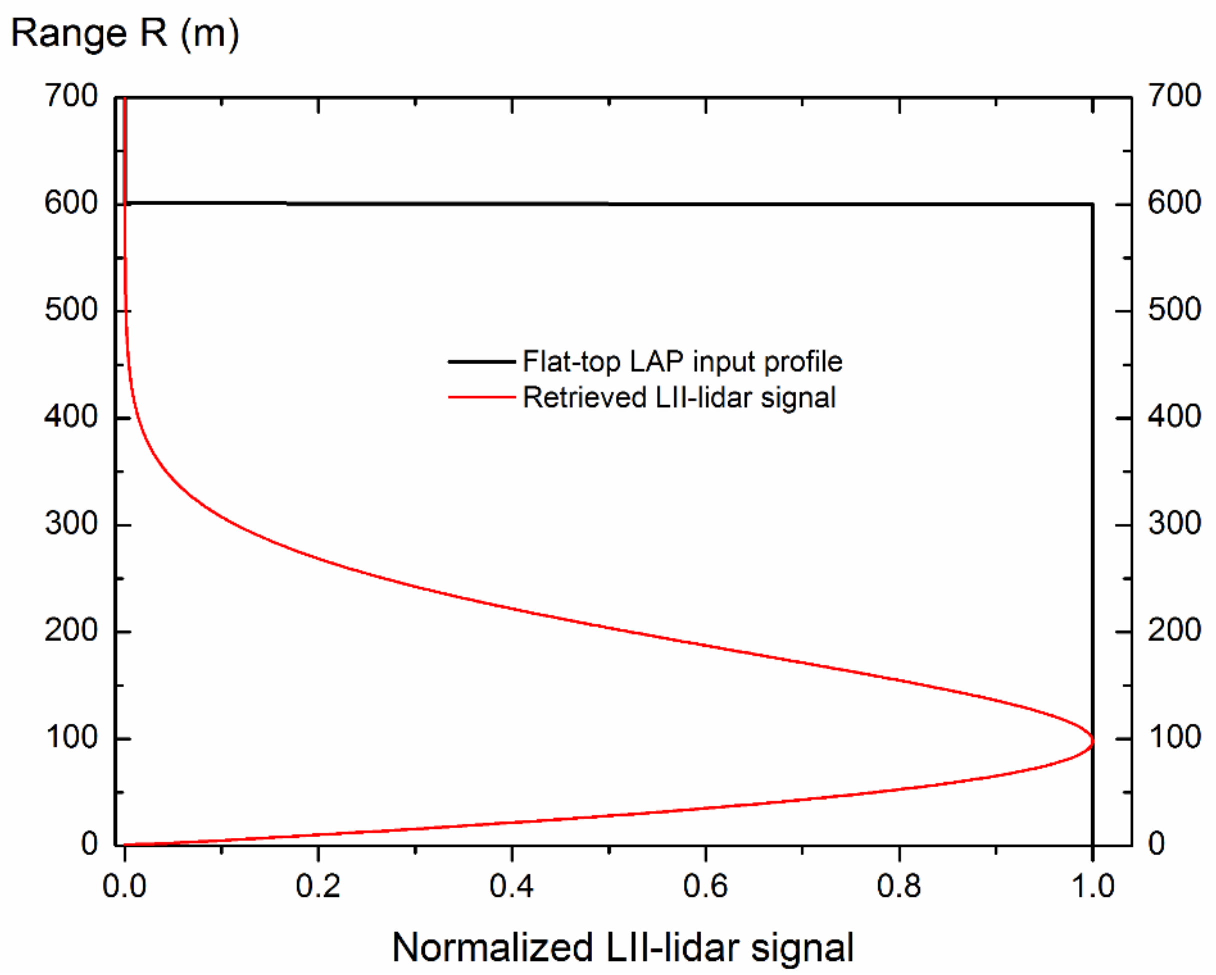

**LAP-peak temperature $T_{max}$ at a range R**

(a) $E_L$ = 150 mJ, beam divergence: $\theta_L$ = 0.01 mrad (dotted line), $\theta_L$ = 0.07 mrad (full line), $\theta_L$ = 0.1 mrad (dash-dotted line), $\theta_L$ = 0.1 mrad (dashed line), for the following set of laser parameters ($E_L$ = 150 mJ, $d_0$ = 30 mm).

(b) $\lambda_L$ = 1064 nm and $E_L$ = 500 mJ.

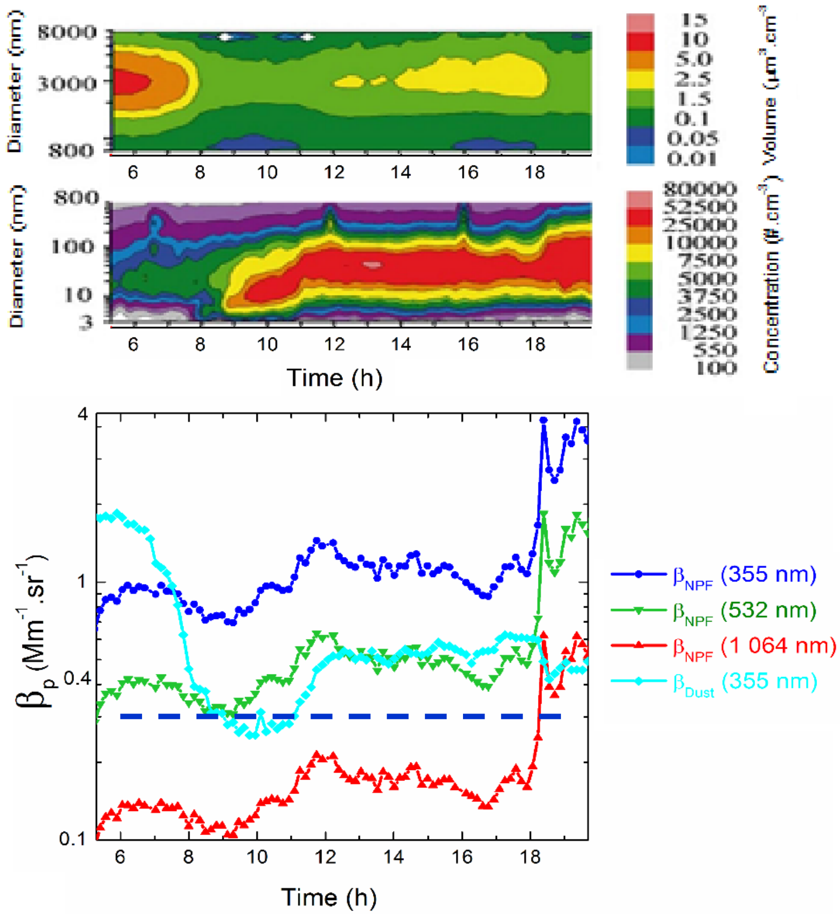
LAP: Diesel soot particles (m = 1.49 + 0.67i, ρ = 1700 kg.m$^{-3}$, $c_p$ = 1900 J.kg$^{-1}$.K$^{-1}$) (Schnaiter et al., 2003)

# LII-Lidar simulation of LAP in the atmosphere




Laser characteristics are $E_L$ = 500 mJ, $\lambda_L$ = 1064 nm, $\theta_L$ = 0.07 mrad and $d_0$ = 30 mm. The LII-radiation is detected at wavelength **$\lambda$ = 633 nm**. LII-lidar signal are normalized to its maximal value.

# Particles nucleation observation with Lidar: sensitivity study




- Newly formed particles are usually assumed to be sulfuric acid (Kulmala et al., Nature Protoc., 2012).
- T-Matrix Numercial simulation of the Backscattering of dust particles.



[1]G. David, A. Miffre et al. OE, 2014, Y. Dupart et al., PNAS 2012.