Physikalisch-Technische Bundesanstalt
Braunschweig and Berlin
National Metrology Institute

RemoteALPHA - New developments in the metrology of environmental radiation monitoring of alpha emitters

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Department 6.3 "*Radiation protection dosimetry*"
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Motivation: Emergency Response Plans

Safety standards for the protection against the dangers arising from the ionising radiation:
The European Directive 2013/59/EURATOM

Article 97

Emergency Management System

-Member states should ensure that account is taken of the fact that emergencies may occur in their territory…
-The emergency management system shall provide for the establishment of emergency response plans…

Article 98

Emergency Preparedness

-Member States shall ensure that emergency response plans are established in advance for the various types of emergencies…
-Member States shall ensure that emergency response plans are tested and revised at regular intervals…

Article 99

International Cooperation

Member States shall cooperate with other Member States and with third countries in addressing possible emergencies on its territory which may affect other Member States or third countries…

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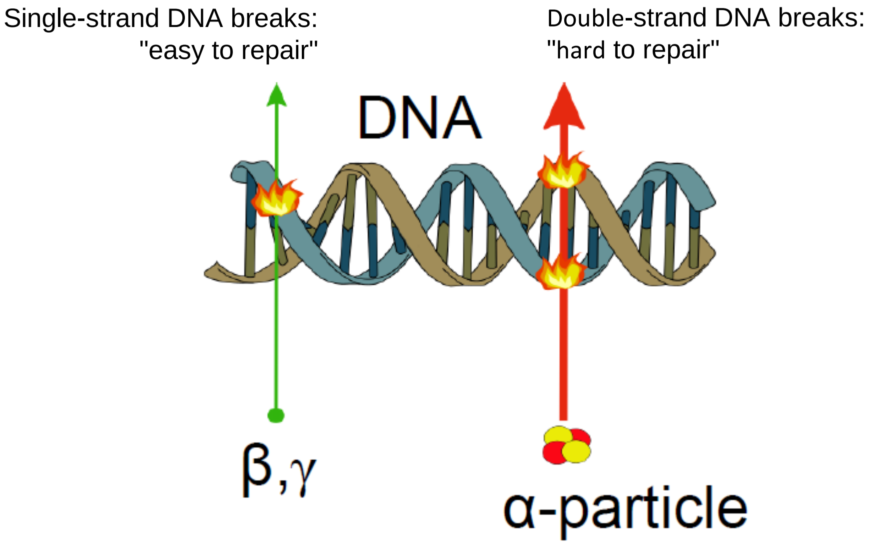
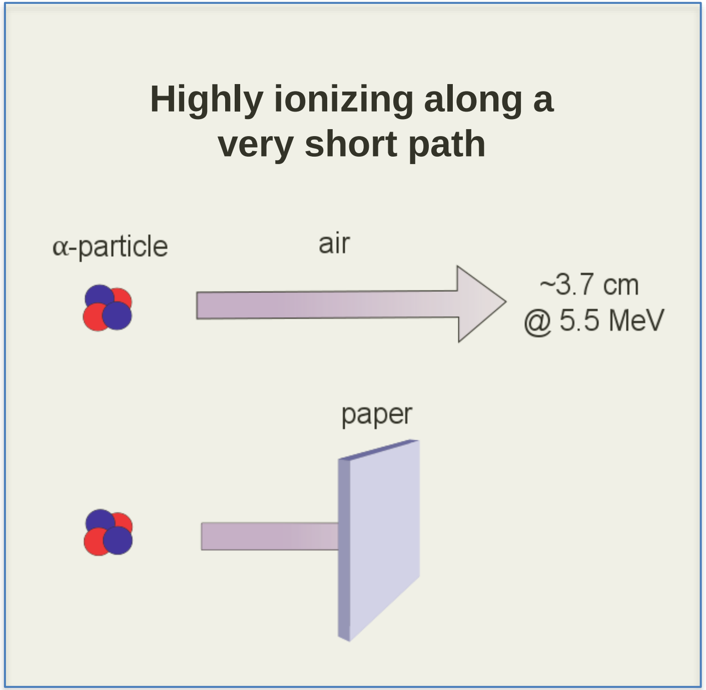
Possible radiological emergency:
Accidental or deliberate dispersion of alpha emitting radionuclides in the environment

Alpha Particles



DNA breaks caused by alpha, beta and gamma radiation.



Alpha Particles. Close Proximity Detection

http://www.argonelectronics.com/blog/the-value-of-applied-learning-for-radiation-safety-training

Traditional detection methods (proportional counter, scintillator counter, PIPS detectors) are:

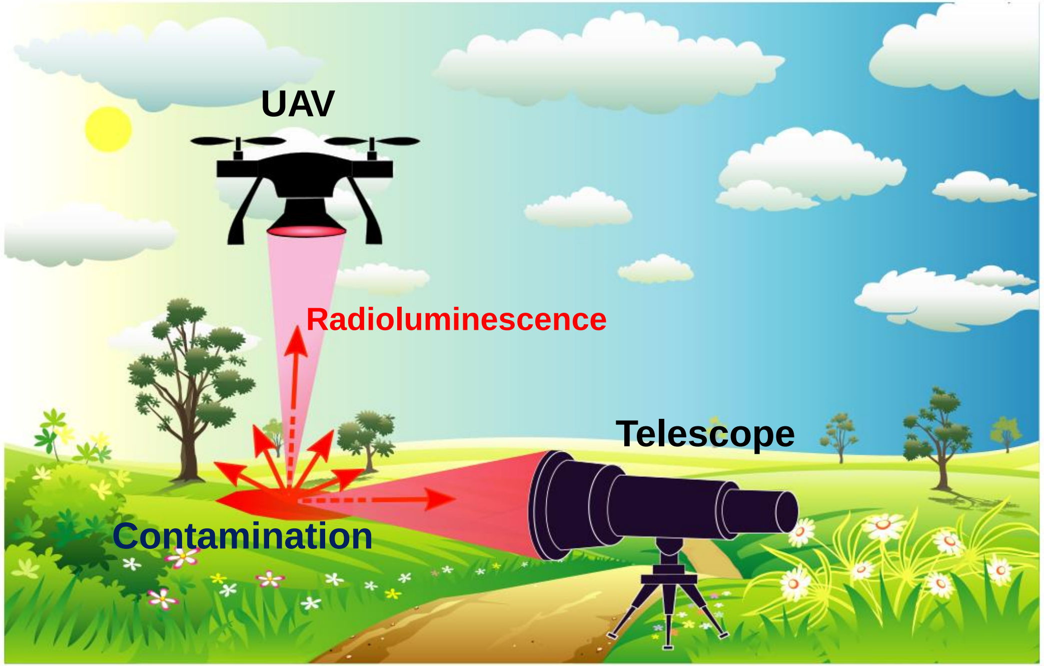
time consuming and tedious,

involve **scanning very close to the surface** of the contaminated area,

require the use **personal protective equipment**,

Expose the personel to other hazards and risks (other types of radiation, fire, etc.).

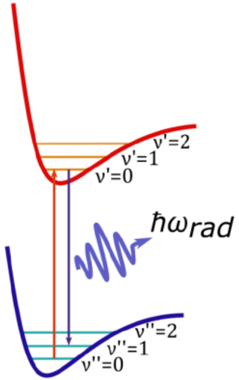
Motivation: Remote detection of alpha particles



Concept of remote detection of alpha particles.

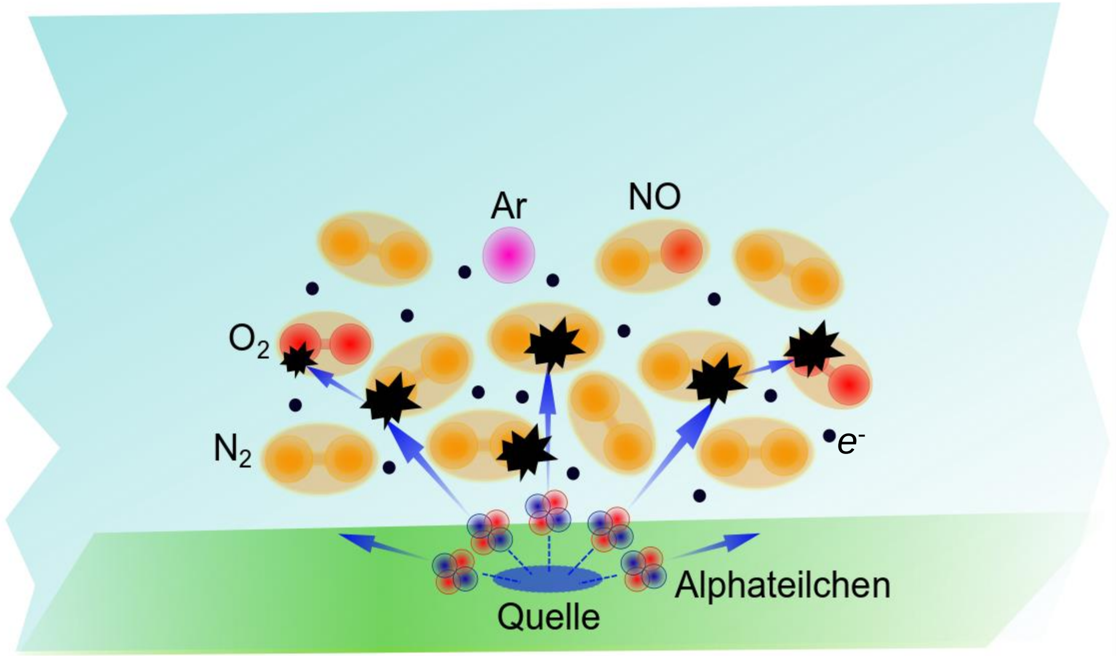
<u>Advantages:</u>

- Operators are kept out of the radiation field,
- Efficient scanning of large areas.



Use of optical transitions in gas molecules: **radioluminescence**

Radioluminescence at a glance



Schematic representation of air ionization by α-particles.

High-energy alpha particles ionize air (predominantly molecular nitrogen).

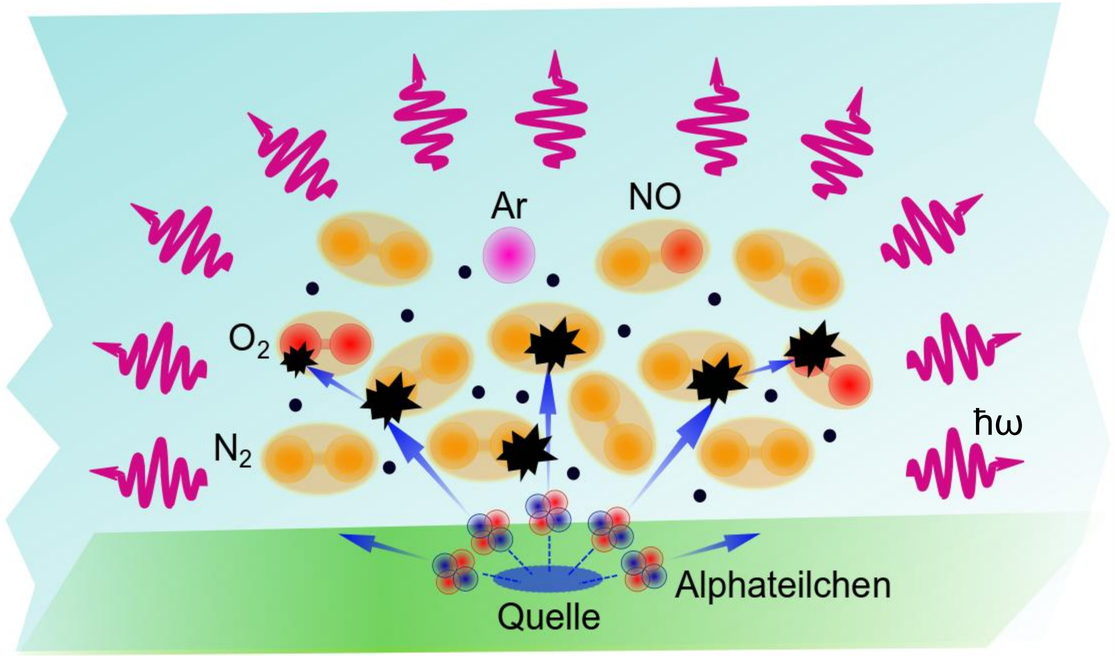
Secondary electrons excite the air molecules, e.g.,

$$e^- + \mathrm{N}_2\left(\mathrm{X}^1\Sigma_g^+\right) \rightarrow \mathrm{N}_2^*(\mathrm{C}^3\Pi_u) + e^-$$

$$e^- + \mathrm{N}_2^+\left(\mathrm{X}^2\Sigma_g^+\right) \rightarrow \mathrm{N}_2^*(\mathrm{C}^3\Pi_u)$$

$\mathrm{X}^1\Sigma_g^+, \mathrm{C}^3\Pi_u \rightarrow$ Molecular levels

Radioluminescence at a glance



Schematic representation of air ionization by α-particles and radioluminescence.

Air molecules emit fluorescent light (radioluminescence) in the UV range between 200 nm and 400 nm.

<u>Range in air:</u>

α-particles → 0,04 m
UV light → 500 m

EMPIR Project: RemoteALPHA

Remote and real-time optical detection of alpha-emitting radionuclides in the environment

Partners

Messen ▪ Forschen ▪ Wissen

Tampere University

Leibniz Universität Hannover

Budapest Főváros Kormányhivatala

Universitat Politècnica de Catalunya BarcelonaTech

IFIN-HH

Szent István University

Alfa Rift Oy

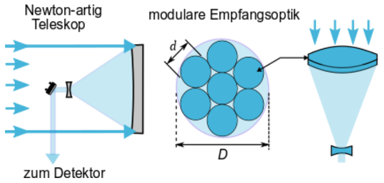
Collaboration:

Australian Government

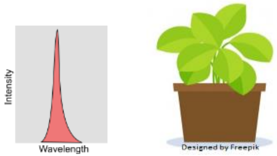
ANSTO

Technical Workpackages

RemoteALPHA: 01.09.2020 - 31.08.2023

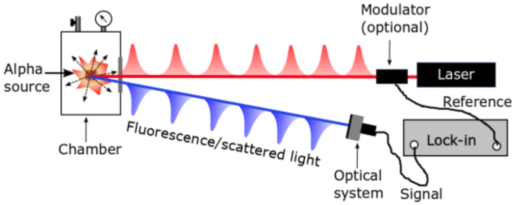


WP1 New instruments for the optical detection of alpha emitters in the environment



WP2 Calibration system for the novel radioluminescence detector systems

WP3 Mapping alpha contamination in the environment using UAVs

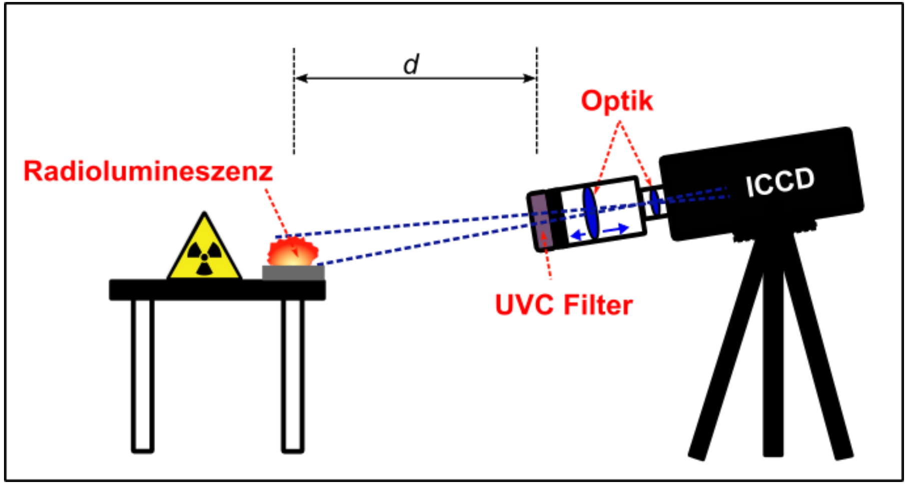


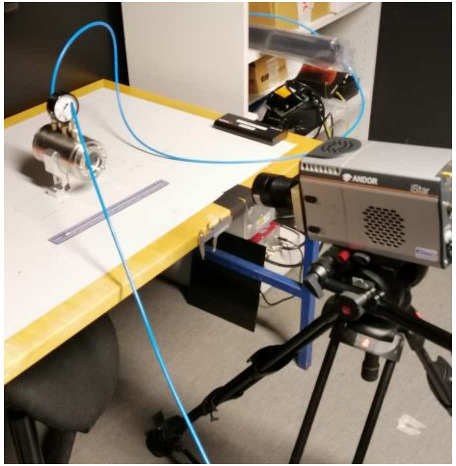
WP4 Feasibility study on laser-based techniques for alpha emitter detection

Imaging of alpha emitters in the UVC (solar-blind) spectral range

Experiments at the University of Tampere (Finland), Research Group of Prof. Juha Toivonen

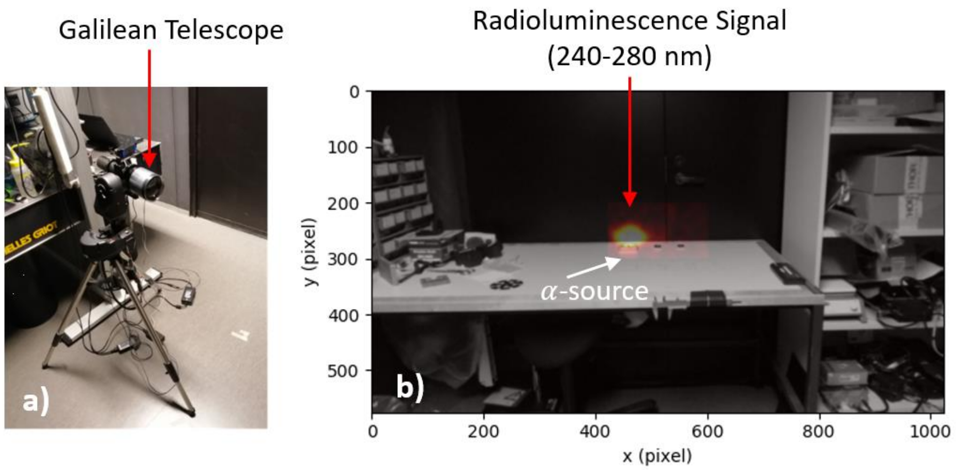
F. S. Krasniqi, T. Kerst, M. Leino, J.-T. Eisheh, H. Toivonen, A. Röttger, J. Toivonen,
Nuclear Inst. and Methods in Physics Research, A **987** (2021) 164821



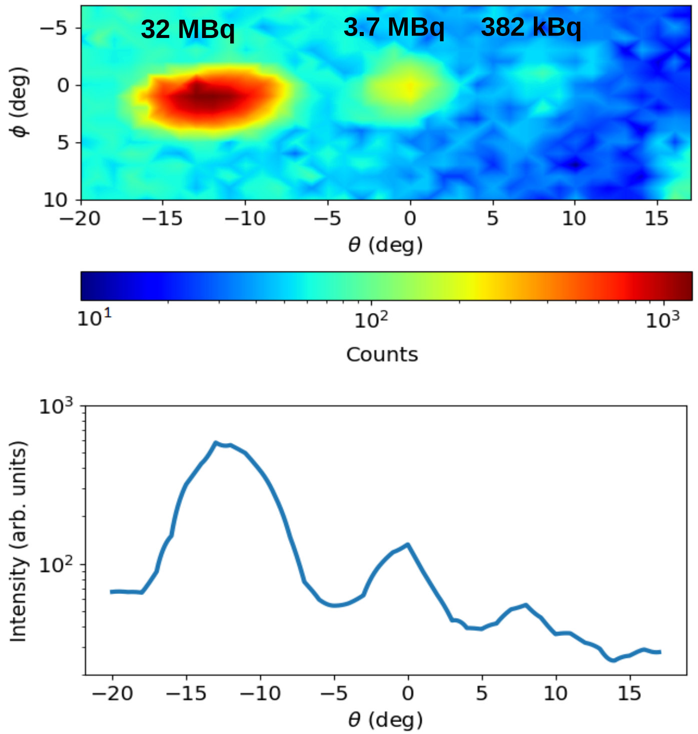


Schematic representation of the UV-C test setup.

UV-C radioluminescence: Detection with telescope and PMT



(a) A photo of the optical system for alpha particle detection. (b) Radioluminescence image of the Am-241 sample (32 MBq) in the UV-C spectral region.

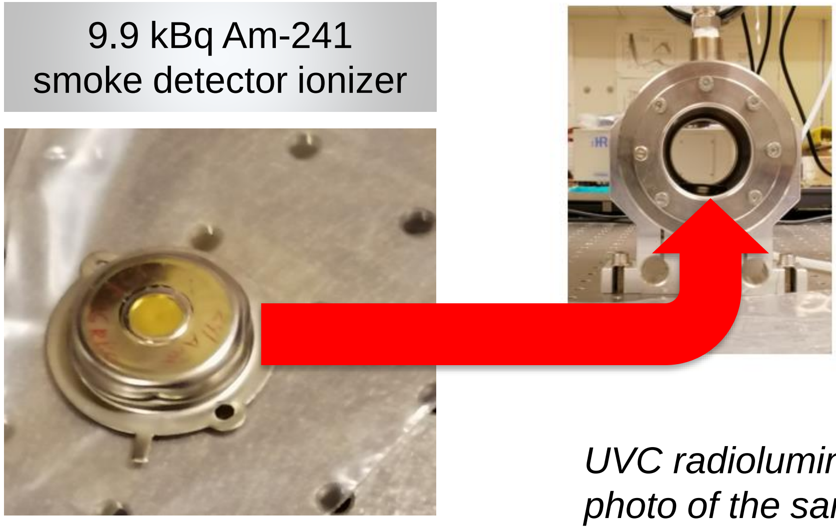


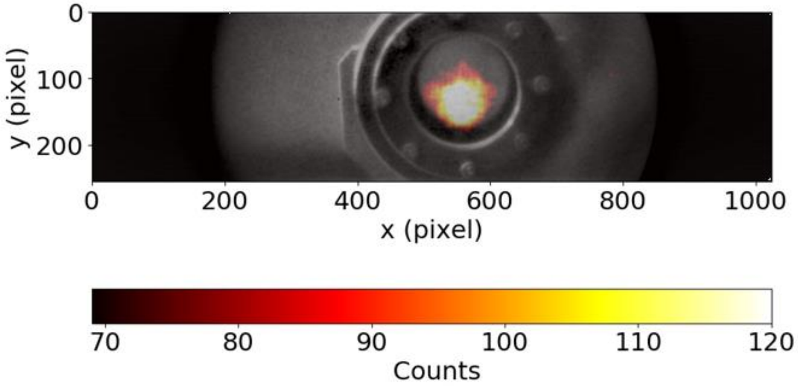
Typical radioluminescence intensity distribution.

UV-C radioluminescence: Amplification with NO

By adding only 3 ppm NO to the air/N_2 amosphere,
up to 500-fold increase of the radioluminescence signal.

9.9 kBq Am-241
smoke detector ionizer

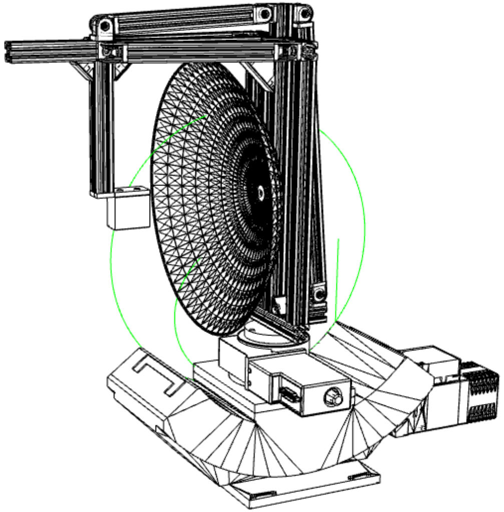


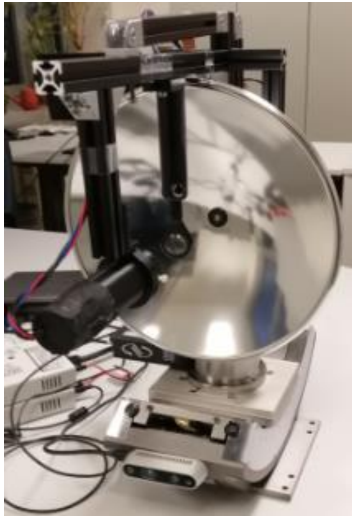


UVC radioluminescence image of a 9.9 kBq source (right) and a photo of the sample in the experimental chamber (left).

Outlook

Optimisation of optical configurations for detection of alpha-induced radioluminescence





Systems based on the reflective optics

Systems based on refractive optics (lenses)

Outlook

Testing of the radioluminscence setup with **environmental samples**

well characterised **alpha-active environmental samples**
concrete, mineral-phase, soil, organic and plant

Acknowledgments

The project 19ENV02 RemoteALPHA has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme.

19ENV02 RemoteALPHA denotes the EMPIR project reference.

Thank you!

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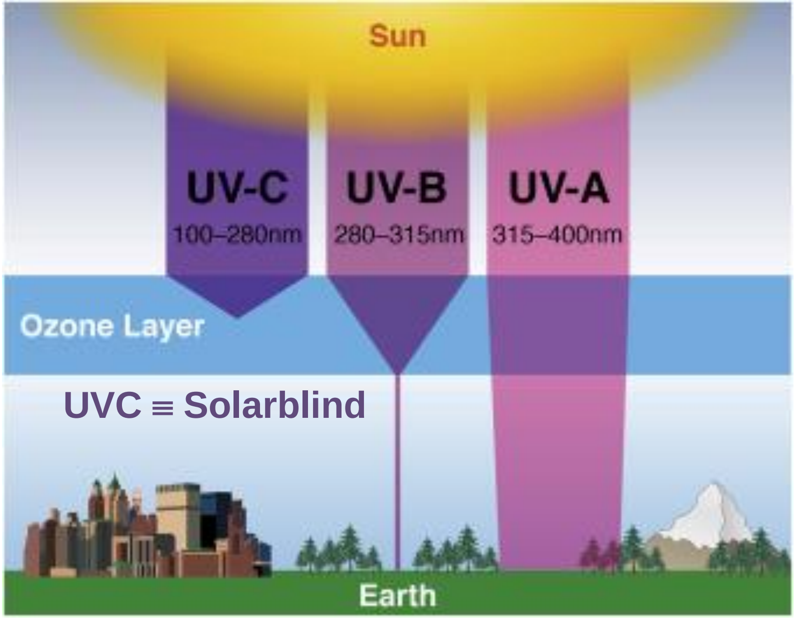
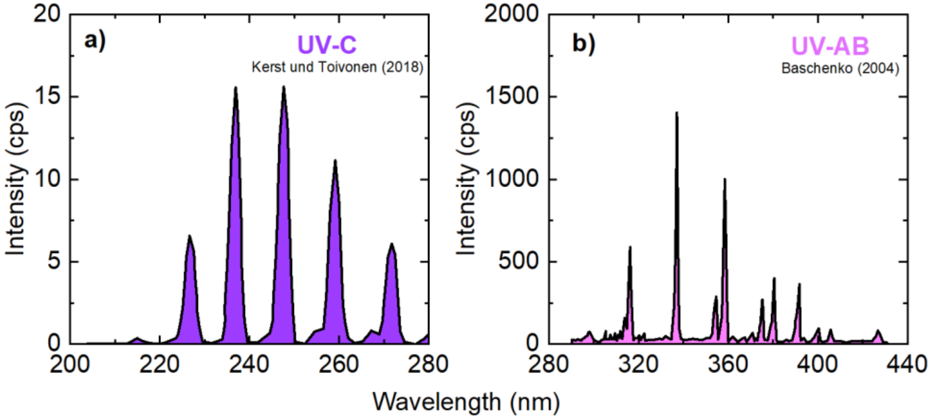
UV-C radioluminescence: interference from sunlight is supressed



D. W. Wilmouth et. al., Green Chemsitry, Ch. 3.3 (2018), https://doi.org/10.1016/B978-0-12-809270-5.00008-X



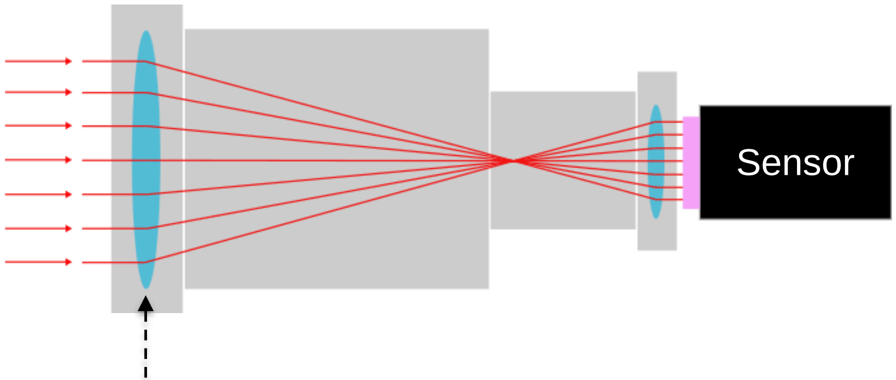
Radioluminescence spectrum

*a) T. Kerst und J. Toivonen, Optics Express **26**, 33764 (2018), und*
*b) S. M. Baschenko, J. Radiol. Prot. **24**, 27 (2004)*

Optimization of the optical system

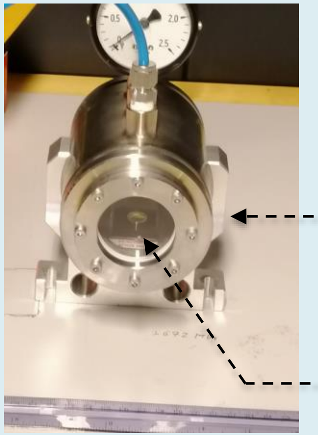
Main optimization parameters:

1) Enlargement of the receiving optics
   (e.g. from ∅ 100 mm to more than 300 mm)
2) Change of the arrangement of the optical
   elements (lenses and mirrors)



Receiving optics

Resesarch on radioluminescence amplification



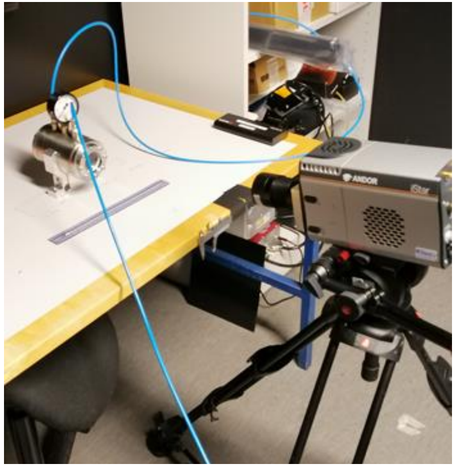
Dependence on gas atmosphere

Chamber with quartz-window

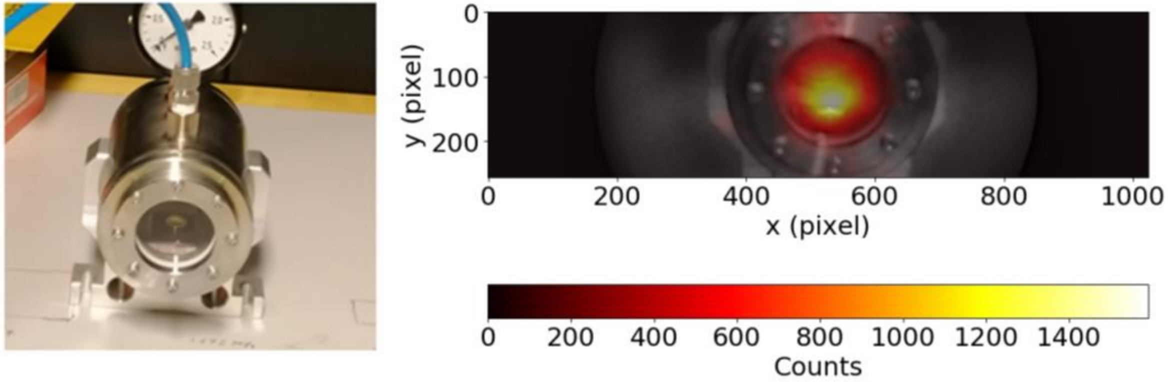
Alphasource

UV-C radioluminescence: Amplification with N_2

Flooding the chamber with N_2, at a rate of 5 L/min
increases radioluminescence by about a hundredfold compared to air.



Experimental setup with CCD

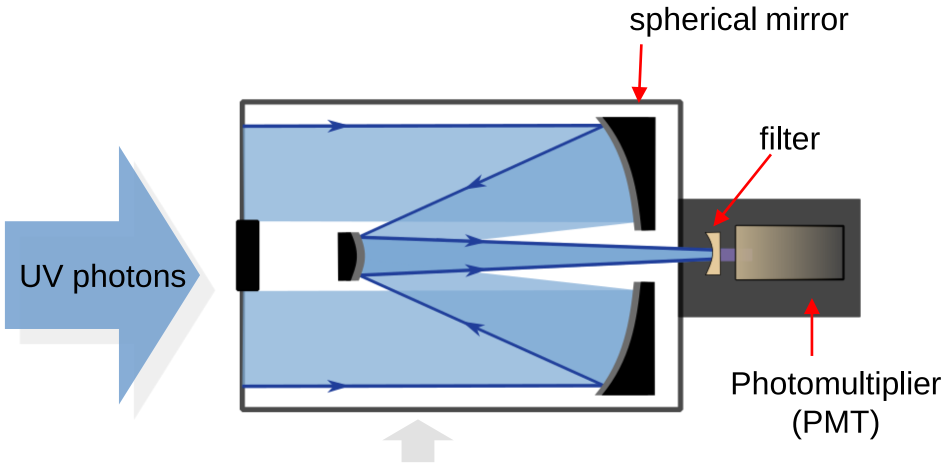


UVC radioluminescence image of a 3.7 MBq source (right) and a photo of the sample in the experimental chamber (left).

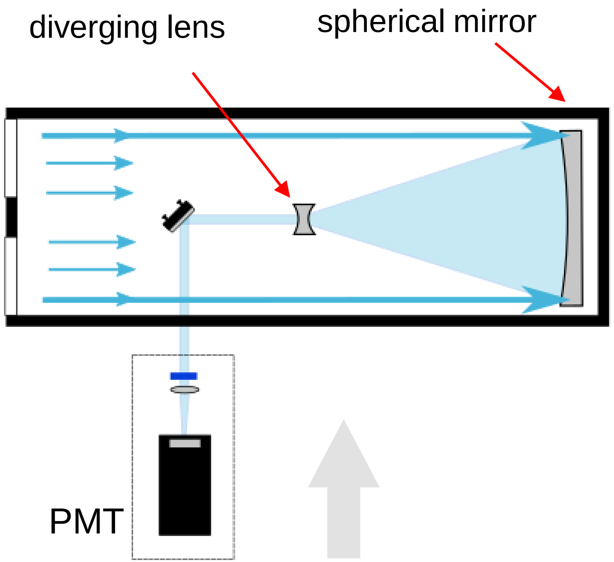
RemoteALPHA: Workpackages

WP1	To develop a new method and instrumentation for the optical detection of alpha particle emitters in the environment by air radioluminescence over a detection range of more than two metres. This includes the development of the first prototype of a mobile-outdoor optical detection system for real-time radioluminescence mapping of alpha sources in the environment.
WP2	To develop and establish a calibration system for the novel-type radioluminescence detector systems. This includes a new metrological infrastructure with a dedicated UV radiance standard, well characterised alpha-active environmental sample (mineral-phase, soil, organic and plant specimen spiked with alpha emitters) and a validated calibration scheme for the remote detection of optical system.
WP3	To extend the optical detection system to an imaging functionality for mapping of alpha contaminations in the environment. This includes the development of an unmanned airborne monitoring system (UAMS) that will integrate the unmanned aerial vehicle (UAV) and the novel alpha-radioluminescence detection system developed in the objective 1 to scan and obtain an image of the contaminated area.
WP4	To prepare and run a feasibility study for a laser-induced fluorescence spectroscopic method for the detection of alpha emitters. This method complements alpha-radioluminescence and, depending on laser parameters such as pulse power, photon wavelength and pulse duration, can enhance the detectable activity limit to below 1 kBq/cm2.

Optical Systems for Remote Alpha-Monitoring



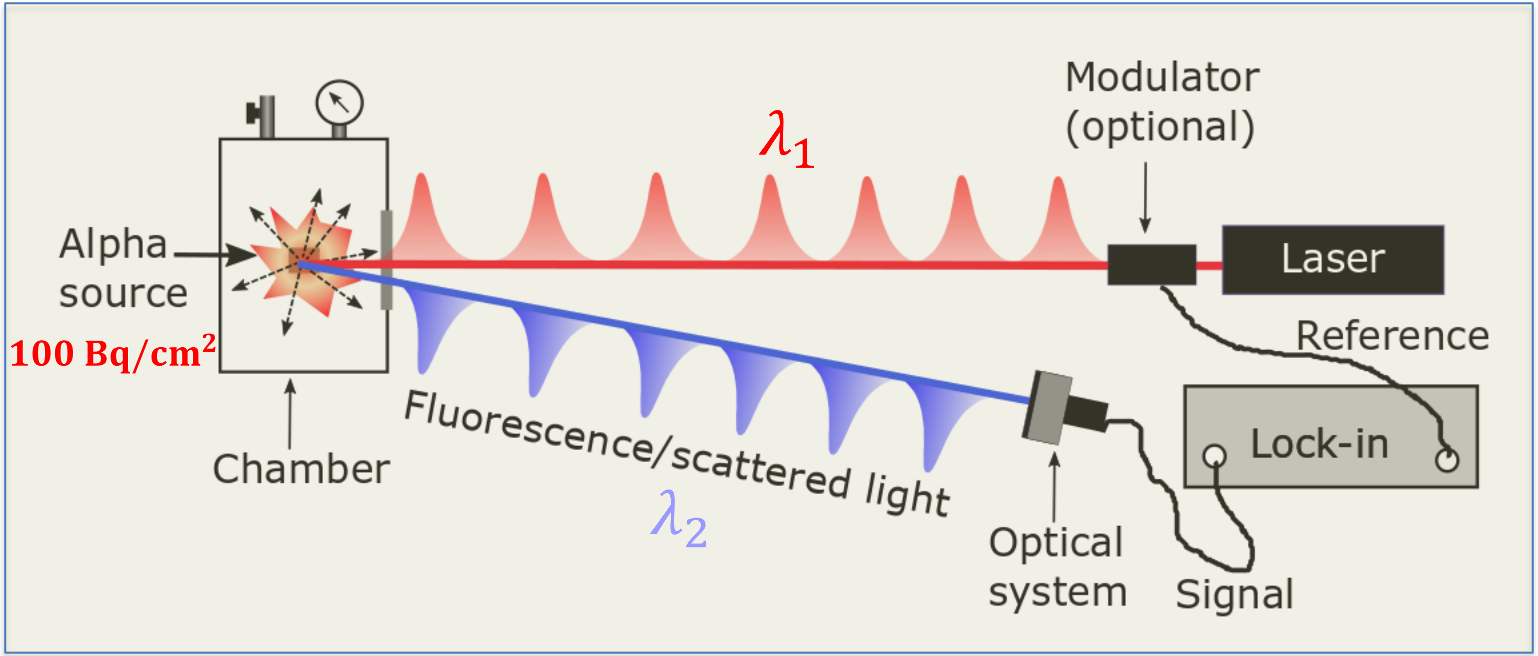
Schematic of a Cassegrain telescope. By folding the light path one can reduce considerably the physical length of the telescope tube.



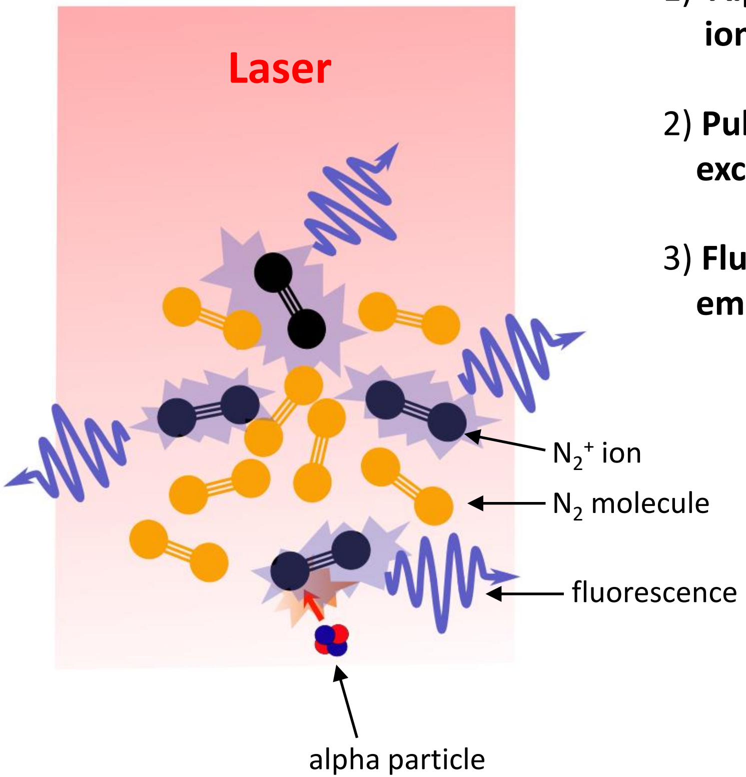
Schematic of a Newtonian-type telescope. Flexible optical system.

Laser Lab Tests: Fluorescence/Raman LIDAR

Supercontnuum fiber laser
Power: ~10 W
Rep rate: 20-320 MHz
Spectral coverage: 400 - 2400 nm



Fluorescence/Resonant Raman Method at a Glance

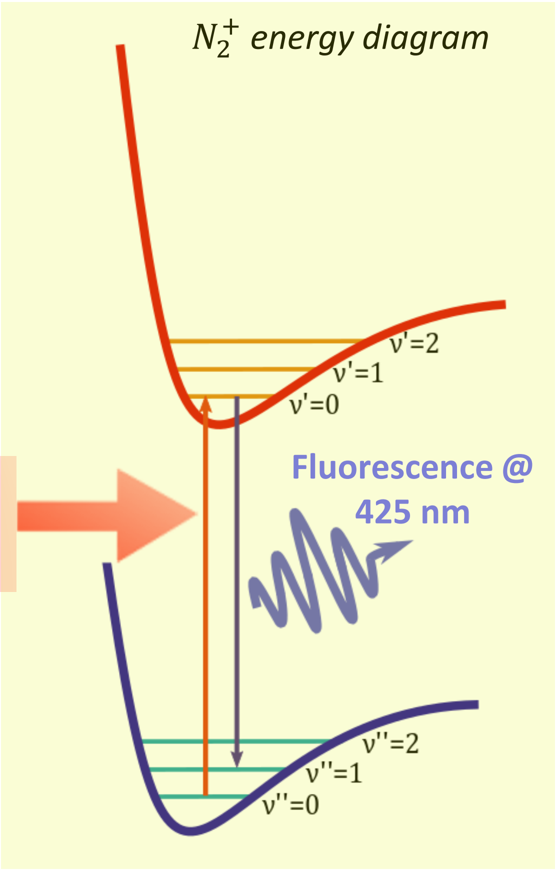


1) **Alpha particles ionize** N_2 molecules.

2) **Pulsed laser excites** N_2^+ ions at λ_{las}.

3) **Fluorescence is emitted at** $\lambda_{emission} \neq \lambda_{las}$

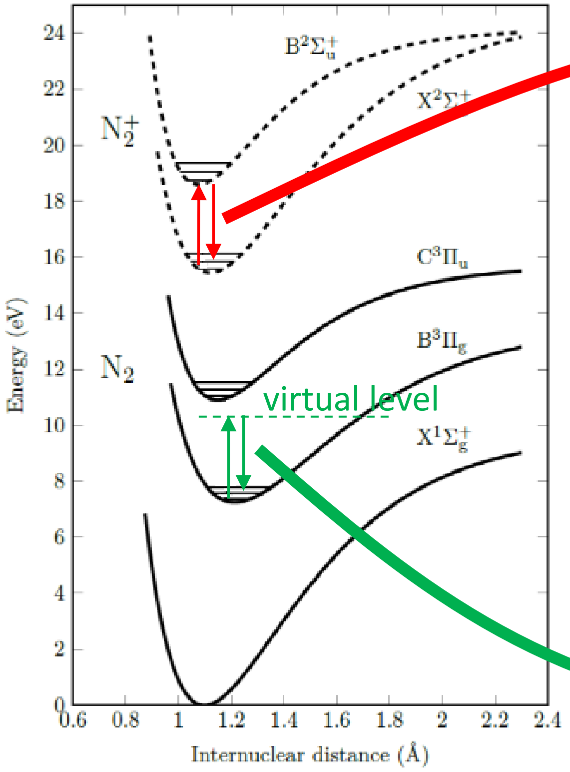
Laser wavelength: 391 nm
Energy per pulse: 500 µJ
Rep. Rate: 10 Hz

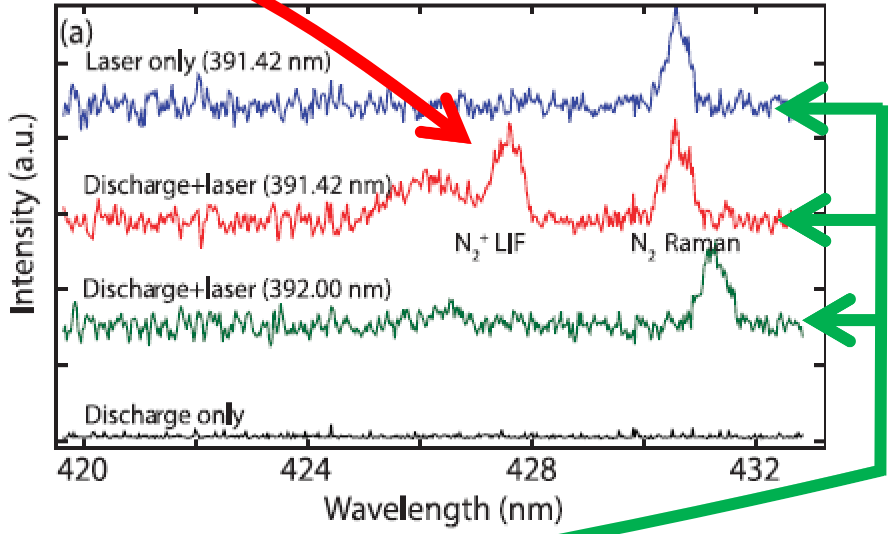


Laser induced fluorescence in ambient air (corona discharge)

Ref.: Konthasinghe *et al.*, Applied Spectroscopy **69**, 1042 (2015)

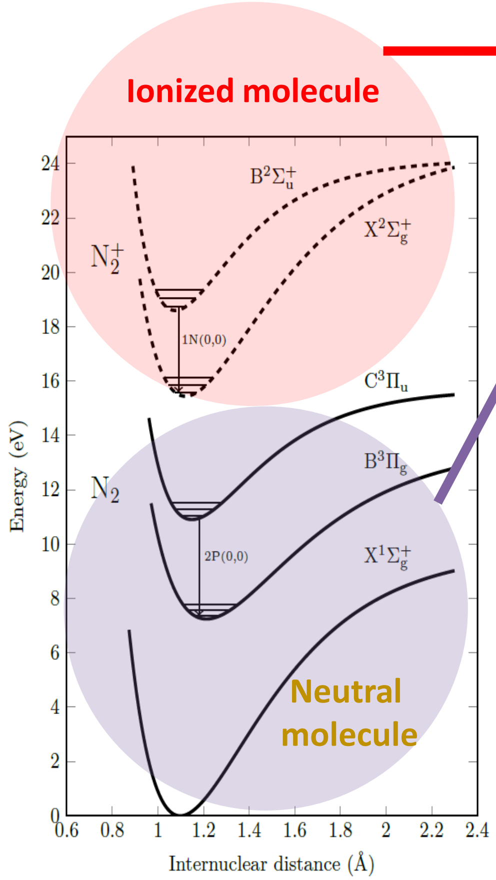
Resonant Raman from N_2^+ (Fluorescence)

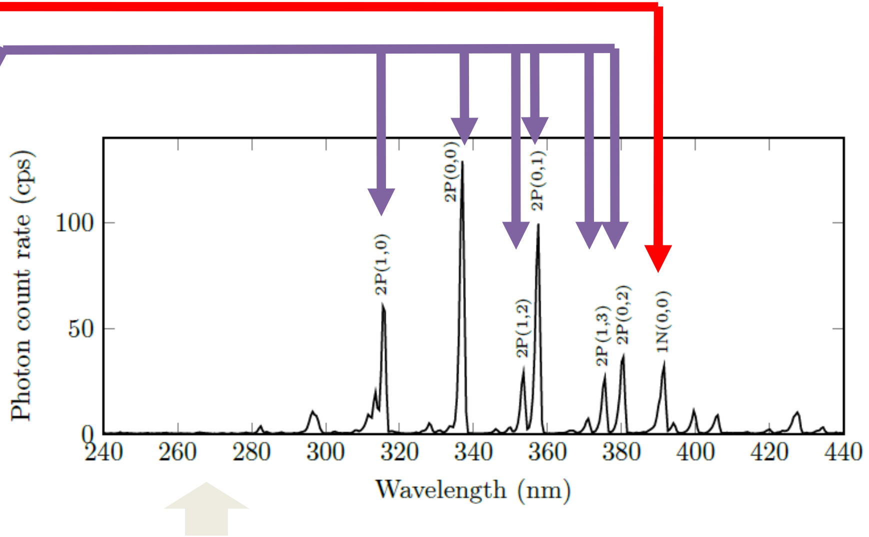




Non-resonant Raman from N_2 (Stokes)

Energy Levels of N_2 Molecule

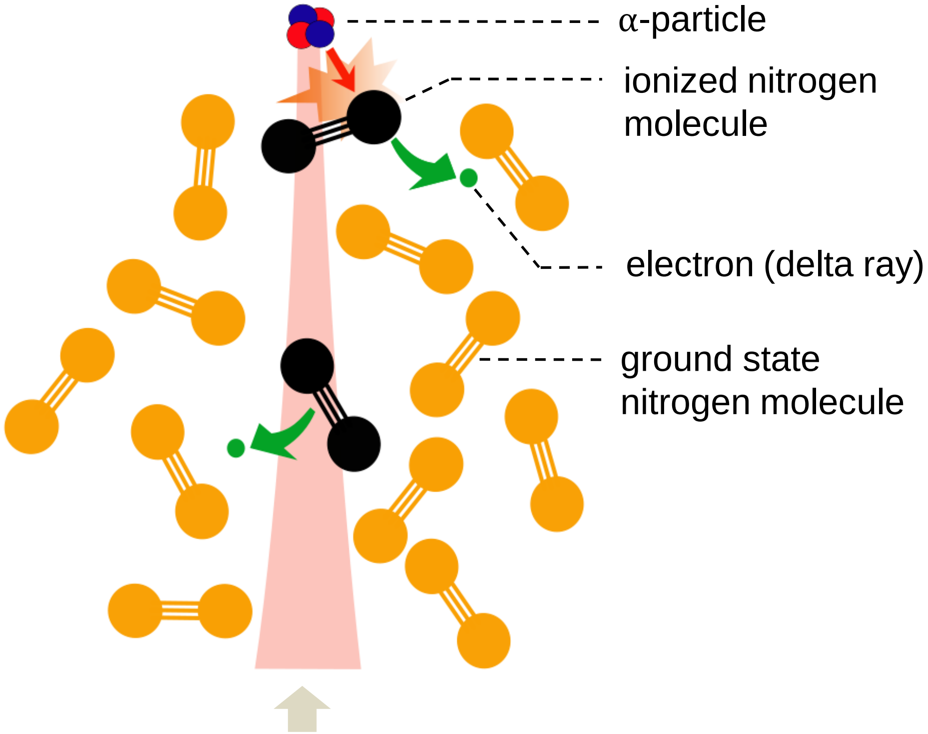




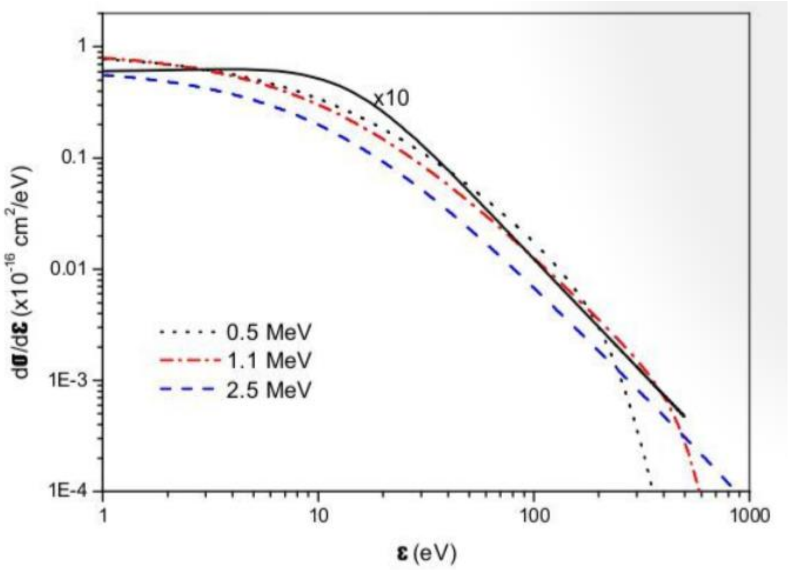
Radioluminescence emission spectrum of air excited by Am-241 (32 MBq) alpha particles. The measurement was performed with a monochromator (Horiba iHR 550) and a low dark count rate photomultiplier tube (PMT) (Perkin Elmer, MP-1982P).

[After Sand, J. (2016). *Alpha Radiation Detection via Radioluminescence of Air.* (Tampere University of Technology. Publication; Vol. **1449**)].

Excitation of Nitrogen



Schematic of nitrogen ionization by α-particles.

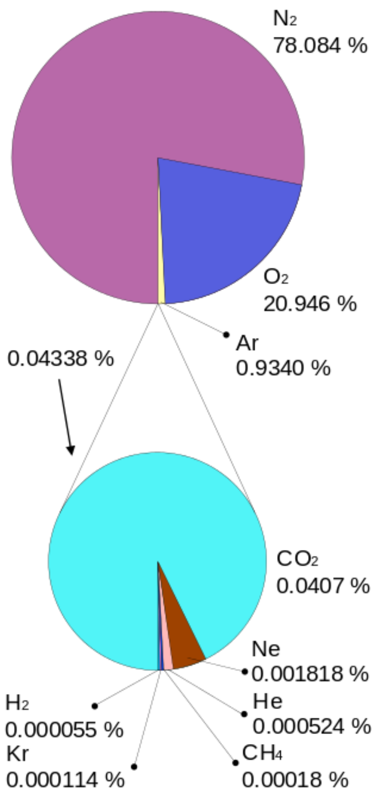


Energy distribution of the electrons produced by α-particles [After: M. M. Farga *et al.*, Nucl. Instrum. Methods Phys. Res. A **597**, 75 (2008)].

Atmosphere of Earth

Major constituents of dry air, by volume

Name	Formula	Volume in %
Nitrogen	N_2	78.084
Oxygen	O_2	20.946
Argon	Ar	0.9340
Carbon dioxide	CO_2	0.041332
Neon	Ne	0.001818
Helium	He	0.000524
Methane	CH_4	0.000187
Krypton	Kr	0.000114
Not included in above dry atmosphere:		
Water vapor	H_2O	0-3%



https://en.wikipedia.org/wiki/Atmosphere_of_Earth

Constituents of the air

Number densities of air constitents:
n_{air} = 2.65×10^{19} molecules/cm^3
n_{N2} = 2.07×10^{19} molecules/cm^3
n_{O2} = 5.55×10^{18} molecules/cm^3
n_{Ar} = 2.48×10^{17} atoms/cm^3
n_{CO2} = 8.75×10^{15} molecules/cm^3

Number density of 1ppm NO:
$n_{NO}=2.47\times10^{13}$ molecules/cm^3

Molar mass of NO:
M_{NO}=14.007 g/mol+15.999 g/mol=30.006 g/mol

Separation between air molecules

$$d_{air} = \left(\frac{3}{4\pi\, n_{air}} \right)^{1/3}$$

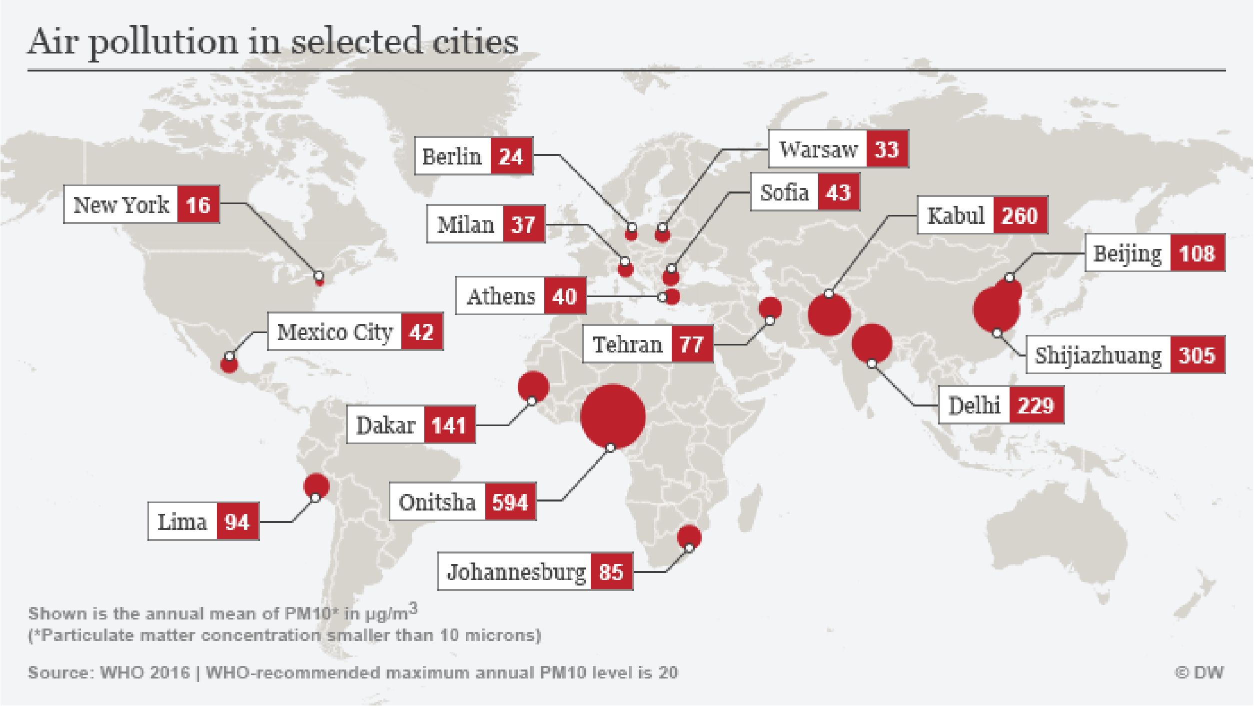
d_{air}~2 nm

Radius of a molecule r~2x Van der Waals radius
For N_2, r~2 x 0.155 nm=0.31 nm

Air molecules occupy only (r/d_{air})^3=0.4% of the volue they are spread.

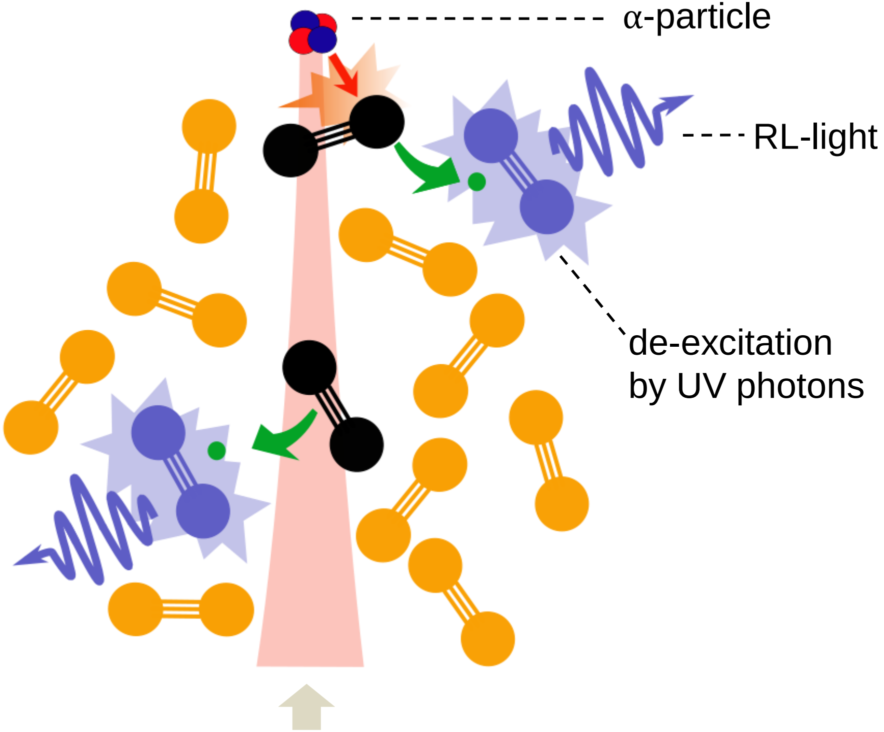
1 ppm NO =1230 µg/m^3 =1.23×10^{-9} g/cm^3
After http://www.euro.who.int/__data/assets/pdf_file/0017/123083/AQG2ndEd_7_1nitrogendioxide.pdf

NO$_x$ concentration



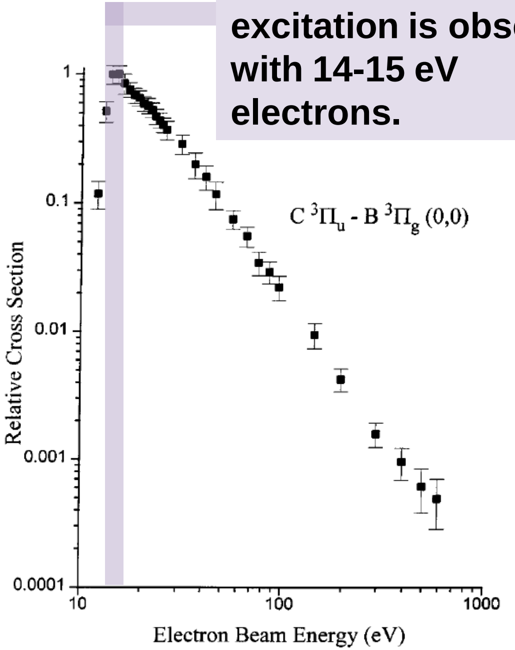
The German city of Stuttgart, for instance, has recorded levels of 82 micrograms of NO2 per cubic meter of air — that's twice as much as the allowed threshold of 40 micrograms. [After https://www.dw.com/en/eu-takes-germany-to-court-over-air-pollution/a-42351552]

Excitation of Nitrogen



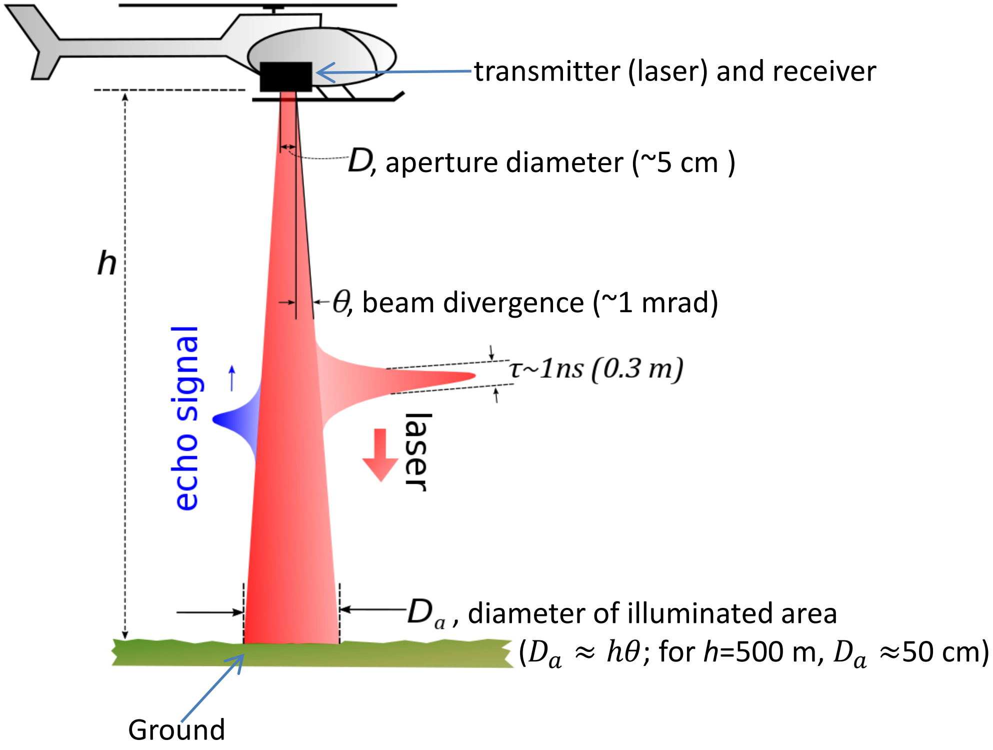
Schematic of nitrogen ionization by α-particles.

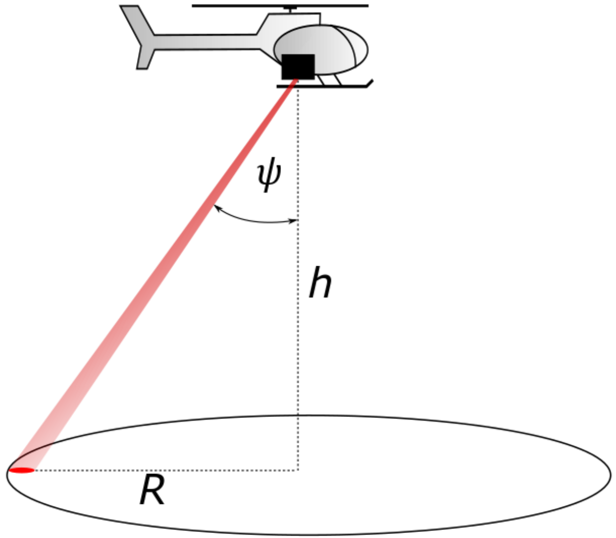
Most effective RL excitation is observed with 14-15 eV electrons.



Relative excitation cross section for the $C^3\Pi_u \rightarrow B^3\Pi_g$ band in N_2. After J. T. Fons *et al.*, Physical Review A **53**, 2239 (1996).

Remote Detection of Alpha Particles: Resonant Raman LIDAR





$R \cong h \tan\psi$

(E. g., for h=500 m and ψ=20 deg, $R \approx$ 182 m)