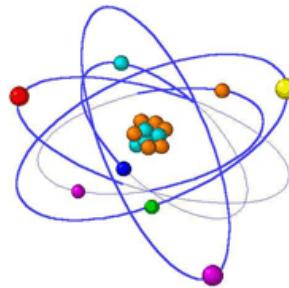


Radioisotope and Radiation Applications (FS2013)



PROTRAC Facility at PSI (Week 7c, Seminar)

Pavel Frajtag

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PROTRAC Facility at PSI: Outline

- ❑ Research Departments and Labs at PSI
- ❑ The Lab of Radiochemistry and Environmental Chemistry
- ❑ The PROTRAC Facility
 - Motivation
 - Production and Delivery System
 - Detection System
 - An Application of PROTRAC
- ❑ The Center for Radiopharmaceutical Sciences
- ❑ Literature / WWW-References

Research Departments and Labs at PSI (1)

PSI is organized into **5 research** and **2 service** departments consisting of laboratories:

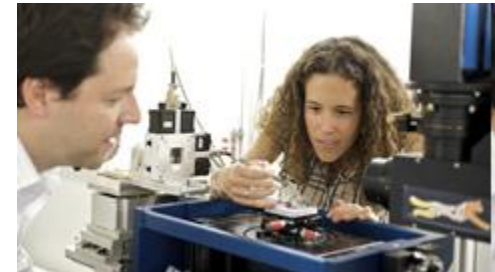
❑ **Research with Neutrons und Muons (NUM)**

- Particle Physics
- Neutron Scattering
- Spallation Neutron Source
- Muon Spin Spectroscopy
- Development and Methods



❑ **Synchrotron Radiation and Nanotechnology (SYN)**

- Macromolecules and Bioimaging
- Condensed Matter
- Energy and Environment
- Micro- and Nanotechnology



❑ **General Energy (ENE)**

- Bioenergy and Catalysis
- Solar Technology
- Combustion Research
- Electrochemistry
- Atmospheric Chemistry
- Energy Systems Analysis ($\frac{1}{2}$)
- Energy and Environment



Research Departments and Labs at PSI (2)

❑ Nuclear Energy and Safety (NES)

- Reactor Physics and Systems Behaviour (LRS)
- Thermal Hydraulics (LTH)
- Hot Laboratory (AHL)
- Waste Management (LES)
- Energy Systems Analysis (LEA) (1/2)
- Nuclear Materials (LNM)



❑ Biology and Chemistry (BIO)

- Center for Radiopharmaceutical Sciences (ZRW)
- Biomolecular Research
- Radiochemistry and Environmental Chemistry (LCH)



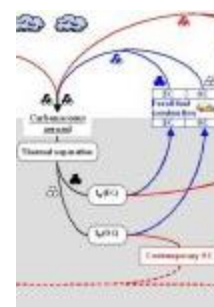
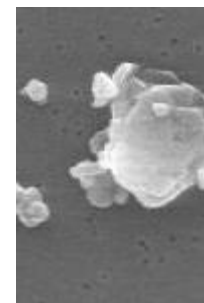
❑ Department Large Research Facilities (GFA)

- Accelerator/Concepts and Development
- Accelerator/Operation and Development
- Technical Support/Co-ordination and Operation

❑ Department Logistics (LOG), provides technical and administrative services

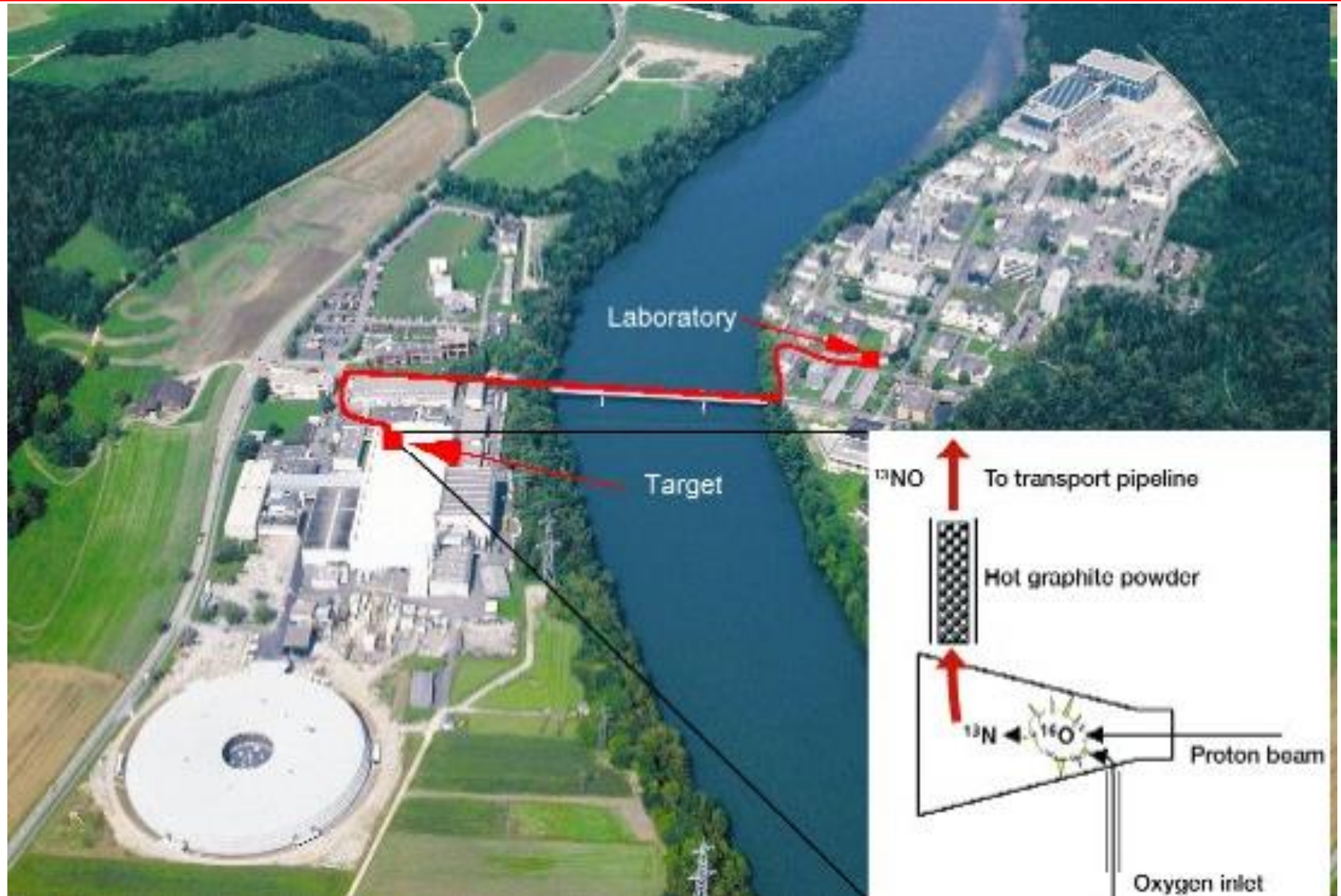
LCH is a joint laboratory of PSI and the University of Bern. Its research is divided in 6 fields:

- ❑ **Heavy elements:** Chemical and nuclear studies of the heaviest elements
- ❑ **Surface chemistry:** Investigation of surface chemistry processes relevant for atmospheric chemistry (**PROTRAC**)
- ❑ **Analytical chemistry:** Past atmosphere and climate reconstructions
- ❑ **Radwaste Analysis:** Development of analytical procedures to characterise nuclear waste samples
- ❑ **Environmental Radionuclides:** Investigation of environmental processes with **natural radioactive tracers**
- ❑ **Radionuclide Development:** Development of **radionuclides for innovative radiopharmaceuticals** ($\frac{1}{2}$ ZRW)



Tb 161 6.91 d β	←
Gd 160 21.86 α 1.5	
Gd 16 3.66 m R	

PROTRAC (1): Overview



PROTRAC (2): Motivation

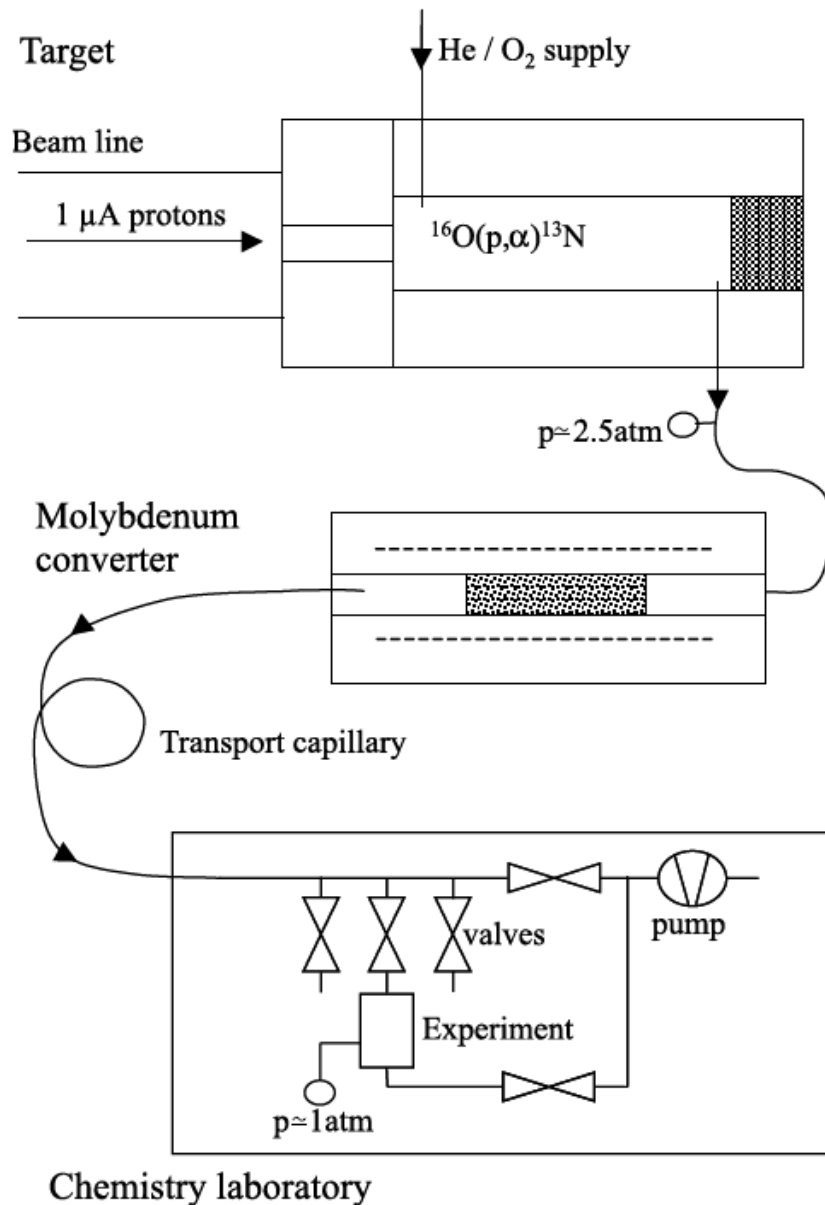
- ❑ A good understanding of **heterogeneous chemistry** is needed in order to adequately describe the cycling of the most important trace constituents, the global and local ozone budget, and the aerosol cloud climate interactions **in the Earth's atmosphere**.
- ❑ Our knowledge on heterogeneous chemistry, which addresses all interactions between the gas-phase and condensed phases such as solid and liquid aerosols, clouds and ground surfaces, is still insufficient.
- ❑ Because of the complexity of the systems involved, adequate results of laboratory experiments are only obtained if the experimental conditions can be compared to the real world.
- ❑ In a reaction involving all molecules covering the surface of aerosol particles of $0.1\ \mu\text{m}$ in diameter typically present in $1\ \text{m}^3$ of air, about 10^{-9} Moles are processed.
- ❑ Therefore **only the most sensitive analytical techniques can be used** in experiments performed at atmospheric concentrations of the trace gas involved.
- ❑ **The use of short-lived radioactive isotopes is an exceptionally ideal tool for studying heterogeneous reactions in the laboratory.** Especially those radionuclides, which decay by emission of γ -rays, are ideal for studying the exchange of molecules with surfaces in complex environments.

PROTRAC (3): Radionuclides produced

10	Ne 20,1797	Ne 16	Ne 17 109,2 ms β^+ 8,0; 13,5... $\beta\alpha$ 4,59; 3,77; 5,12...; $\beta\alpha$ γ 495; 6129*	Ne 18 1,67 s β^+ 3,4... γ 1042...	Ne 19 17,22 s β^+ 2,2... γ (110; 197; 1357)	Ne 20 90,48	Ne 21 0,27
	σ 0,04	2p				σ 0,04	σ 0,7
9	F 18,998403	F 15	F 16	F 17 64,8 s β^+ 1,7 no γ	F 18 109,7 m β^+ 0,6 no γ	F 19 100	F 20 11,0 s β^- 5,4... γ 1634...
	σ 0,0065	0				σ 0,0065	
O 12	O 13 8,58 ms β^+ 16,7... $\beta\alpha$ 1,44; 6,44... γ (4439; 3500...)	O 14 70,59 s β^+ 1,8; 4,1... γ 2318...	O 15 2,03 m β^+ 1,7 no γ	O 16 9,762 β^+ 0,00019	O 17 0,038 β^+ 0,24	O 18 0,200 σ 0,00016	O 19 27,1 s β^- 3,3; 4,7... γ 107; 1357...
2p							
N 11	N 12 11,0 ms β^+ 16,4... γ 4439... $\beta\alpha$ 0,2...	N 13 9,96 m β^+ 1,2 no γ	N 14 99,634 σ 0,000 $\beta\alpha$ 1,8	N 15 0,366 σ 0,00004	N 16 5,9 μ s; 7,13 s β^+ 4,5; 10,4... γ 5129; 7115... $\beta\alpha$ 1,76	N 17 4,17 s β^- 3,2; 8,7... βn 1,17; 0,39... γ 871; 2184; $\beta\alpha$ 1,25; 1,41	N 18 0,63 s β^- 9,4; 11,5... γ 1902; 822; 1602; 2473... $\beta\alpha$ 1,08; 1,41... βn 1,35; 2,46
p							
C 10 19,3 s β^+ 1,9... γ 718; 1022	C 11 20,38 m β^+ 1,0 no γ	C 12 98,90 σ 0,0035	C 13 1,10 σ 0,0014	C 14 5730 a β^- 0,2 no γ	C 15 2,45 s β^- 4,5; 9,8... γ 5298...	C 16 0,747 s β^- 4,7; 7,9... βn 0,79; 1,72	C 17 193 ms β^- 1,62... γ 1375; 1849; 1906...
B 9	B 10 19,9 σ 0,5 σ_{th} 3840	B 11 80,1 σ 0,005	B 12 20,20 ms β^- 13,4... γ 4439... $\beta\alpha$ 0,2...	B 13 17,33 ms β^- 13,4... γ 3884 βn 3,6; 2,4...	B 14 13,8 ms β^- 14,0... γ 6090; 6730 βn	B 15 10,4 ms β^- 1,77; 3,20...	
p							
Be 8	Be 9 100 σ 0,008	Be 10 1,6 $\cdot 10^6$ a β^- 0,6 no γ	Be 11 13,8 s β^- 11,5... γ 2125; 6791... $\beta\alpha$ 0,77...	Be 12 23,6 ms β^- 11,7... βn		Be 14 4,35 ms β^- 0,8; 3,02; 3,52...; 62n γ 3529; 3580*	
2s							
Li 7 92,5 σ 0,045	Li 8 840,3 ms β^- 12,5 $\beta\alpha$ 1,6	Li 9 178,3 ms β^- 13,6... βn 0,7... $\beta\alpha$	Li 10	Li 11 8,5 ms β^- 18,5; 20,4 γ 3366; 320... βn ; $\beta 2n$; $\beta 3n$; $\beta\alpha$; βt			
n							

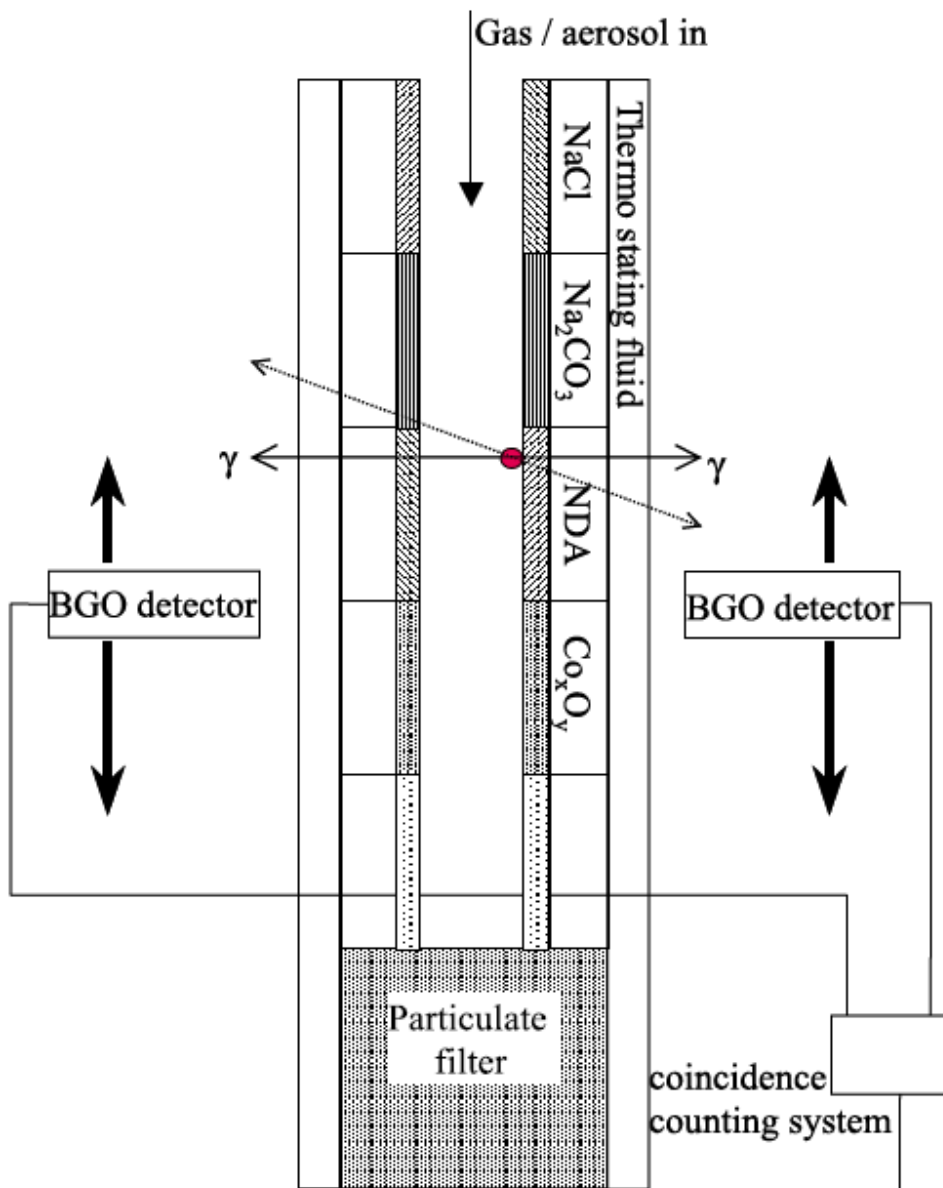
- PROTRAC stands for: **production of tracers for atmospheric chemistry.**
- The aim of the PROTRAC project is to provide the short-lived radioactive isotopes ^{13}N , ^{11}C and ^{15}O to the PSI laboratories for experiments in atmospheric chemistry. To study interactions between gas molecules and surface highly sensitive techniques are used which employ radioactive-labelled molecules. The isotopes are produced by irradiation of a gas stream in a target by a proton beam.
- The short half-life of ^{13}N , ^{11}C and ^{15}O results in a high activity and therewith **high sensitivity.**

PROTRAC (4): Production and Delivery System



- The radionuclides are produced by bombarding a He/O₂ gas target with protons in the energy range between 10 and 15 MeV and with beam intensity from 1 µA to 10 µA. Under these conditions:
 - dominantly the radioisotope ¹³N ($T_{1/2}=9.96\text{min}$) is made via $^{16}\text{O}(p,\alpha)^{13}\text{N}$ reactions,
 - as a byproduct also ¹¹C ($T_{1/2}=20.38\text{min}$) appears from $^{16}\text{O}(p,\alpha p n)^{11}\text{C}$ reactions,
 - and the radioisotope ¹⁵O ($T_{1/2}=2.03\text{min}$) can be produced by $^{16}\text{O}(p,pn)^{15}\text{O}$ reactions.
- All radionuclides are proton deficient and decay by positron emission, which, after annihilation with electrons, leads to a **clear signature of two 511 keV gammas**.
- Under the radiation chemistry in the gas target ¹³N rapidly forms highly oxidized and reactive forms of nitrogen oxides, which are converted to ¹³NO over molybdenum at 500°C.
- In a polyethylene tube of 0.2cm in diameter the ¹³NO is transported over 80m to the laboratory.
- In the laboratory the gas mixture coming from the target containing ¹³NO passes around a NaI detector which determines the amount of ¹³N-labelled molecules on-line, before the radiotracer is distributed to the experiments.

PROTRAC (5): Detection System



□ Denuder and detection system:

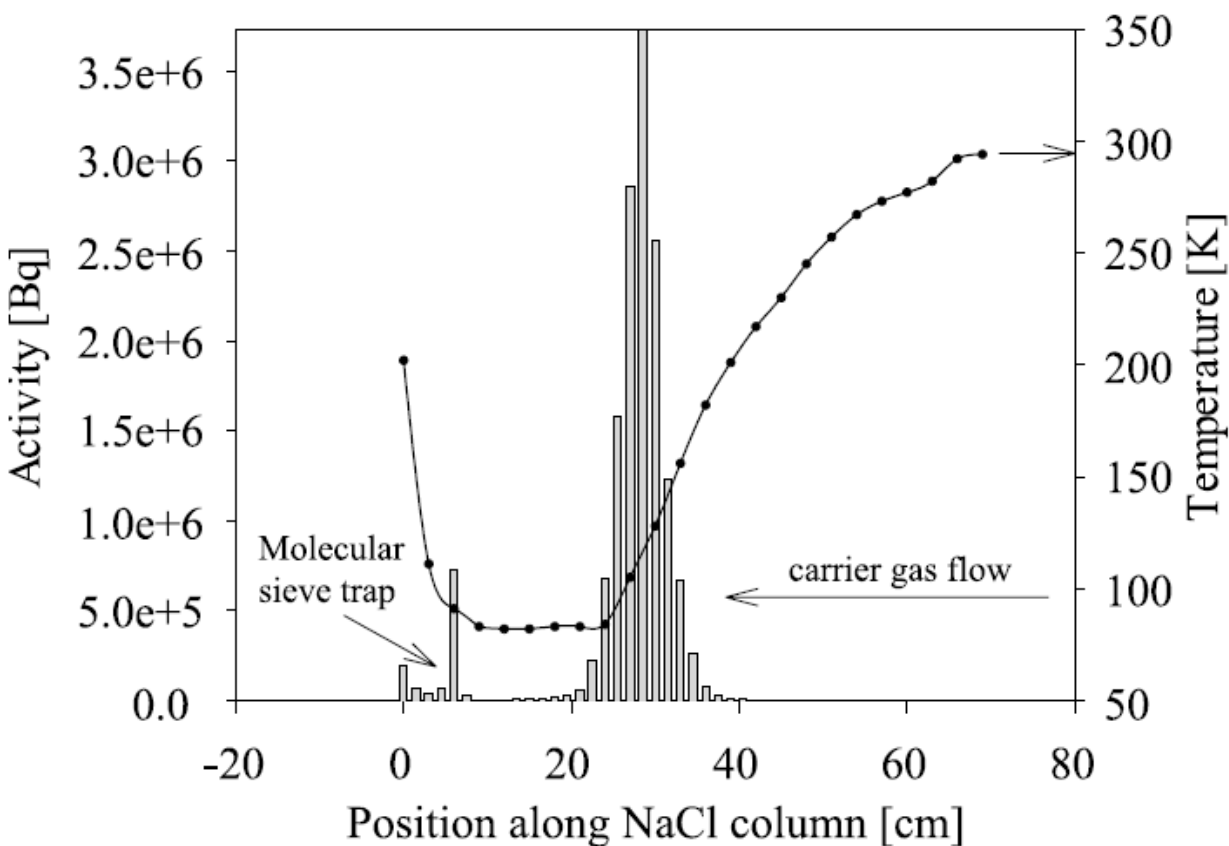
- **gas-phase species** (NO, NO₂, ...) **are absorbed** on the specifically coated denuder plates by diffusion,
- **particles penetrate** with nearly 100% efficiency **to the particle filter**,
- the coincident counting configuration allows quasi-online measurement of the activity distribution on the denuder plates, or on chromatographic columns.

□ BGO = Bismuth germanate (Bi₄Ge₃O₁₂) is an inorganic chemical compound that forms cubic crystals. It is a scintillator (8500 photons per MeV gamma radiation) and is often used in PET detectors.

□ NDA (*n*-(1-naphtyl)diethylenediamine-hydrochloride) is a compound that reacting with NO₂ produces HNO₂ (nitrous acid).

PROTRAC (6)/Application: Adsorption of NO_2 on NaCl

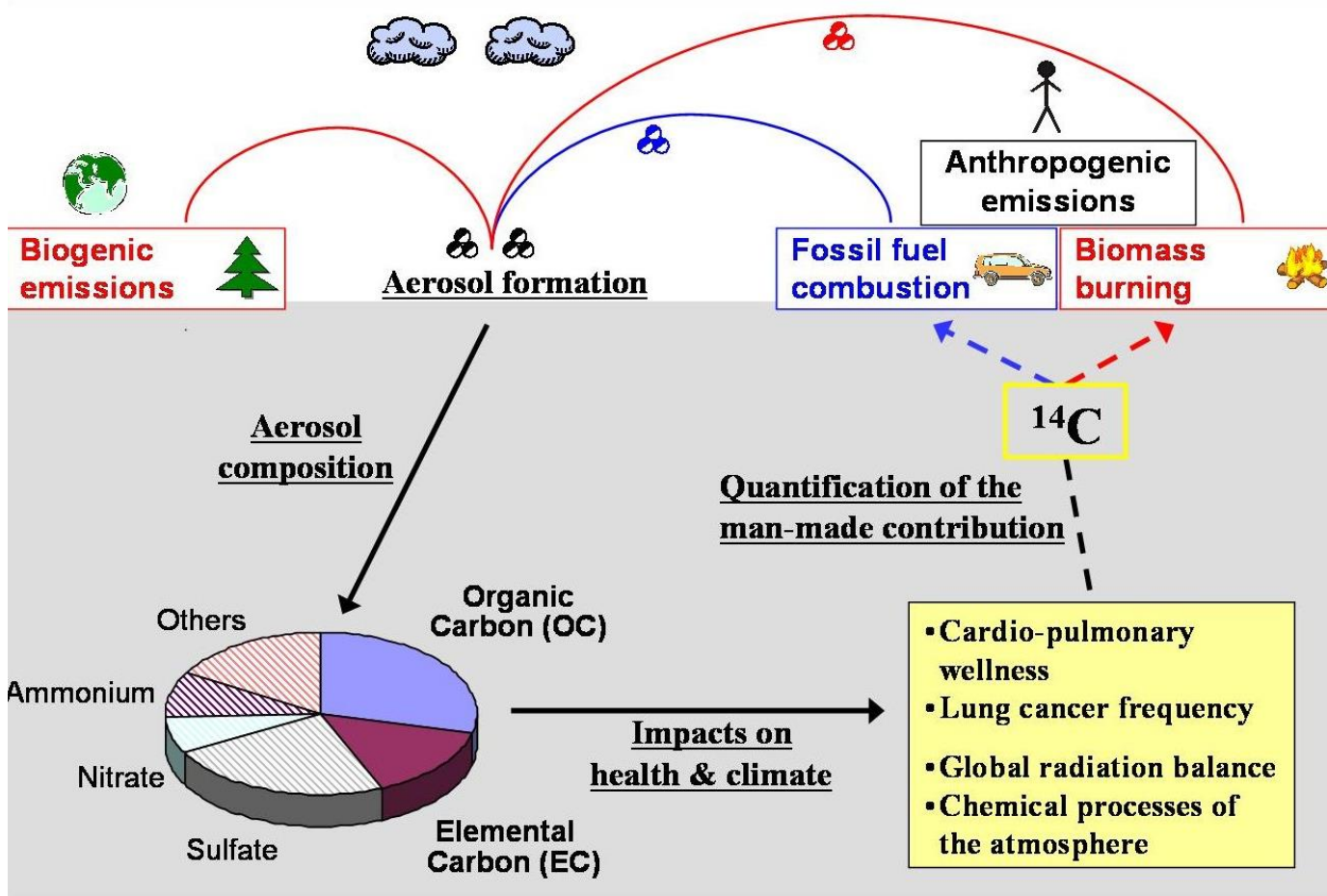
Motivation: In the atmosphere, NaCl is the main constituent of sea salt aerosol generated via sea spray on the oceans and representing a very important aerosol material of the troposphere. Heterogeneous processes involving nitrogen oxides are slowly depleting chloride in the sea salt aerosol by forming nitrates on the aerosol and HCl in the gas-phase.



- In a thermochromatography experiment, a quartz column coated with NaCl was exposed to a flow containing $^{13}\text{NO}_2$ in a temperature gradient (solid line, right axis), with the gas flowing from the warm to the cold end. After the experiment, the activity distribution was measured (bars, left axis). A molecular sieve trap at the end of the coating was used to trap all ^{13}N species not retained in the column.
- The symmetric peak shown in the Figure confirmed that reversible adsorption was the main process governing migration of $^{13}\text{NO}_2$ along the NaCl surface.
- An adsorption enthalpy of 28 kJmol^{-1} was derived from this experiment and it also allowed estimating an upper limit to the reaction probability of NO_2 with NaCl.

Environmental Radionuclides Group of LCH

- Their objective: Source apportionment of carbonaceous aerosols with radiocarbon measurements, i.e.:
 - Apportionment of emission sources of airborne particulate matter
 - Radionuclide dating of environmental archives
 - Development of techniques for radiocarbon analysis
- One of their experimental methods: Accelerator Mass Spectrometry



ZRW is sponsored by the three institutions ETH Zurich, PSI (Department of Biology and Chemistry) and USZ (University Hospital Zurich). Its research is orientated towards clinical application and is divided in 7 fields/groups:

- ❑ **Radionuclide Chemistry:** Coordination Chemistry, Radiochemistry
- ❑ **Folate Receptor Targeting:** Development of Folate-Based Radiopharmaceuticals
- ❑ **Development of Radionuclides** for innovative radiopharmaceuticals
- ❑ **Tumor Affine Peptides:** Peptide Development and ^{99m}Tc / ^{188}Re Radiolabelling
- ❑ **Tumor Targeting:** Antibody Engineering, ^{67}Cu Radiolabelling
- ❑ **Molecular Imaging:** PET-Radioligands for the Brain-Receptors and Tumordiagnosis (**ETH**)
- ❑ **Radioligands for clinical Research:** Development of [^{11}C]MHED for heart studies, D- and L- [^{11}C]-Lactate and [^{18}F]Fluoralanserin for brain studies (**USZ**)

Development of Radionuclides

- An important component of innovative radiopharmaceuticals, especially in oncology is the availability of alternative radionuclides with optimal decay properties, which allow to improve diagnostic or therapeutic efficiency. **Currently a variety of accelerator, reactor (or Spallation Neutron Source) and generator produced radionuclides are developed and produced at PSI.** Medical radionuclide must be available with high specific activity and purity. Here the choice of the nuclear reaction and subsequent radiochemical isolation strategy play a key role.
- Localization and tracking of radiopharmaceuticals in vivo is performed by single photon emission computed tomography (SPECT) as well as by positron emission tomography (PET). The therapeutic radionuclides are particle emitters that are able to deposit a high quantity of energy in small volumes via a high "linear energy transfer" (LET). Here β^- , α and also Auger electron emitters are used or considered to be useful for the therapeutic treatment.
- Currently production of several diagnostic and therapeutic radionuclides is evaluated and established at PSI.

The following produced radionuclides are available for pre- and clinical studies:

- ^{64}Cu ($T_{1/2}=12.70$ h) for PET applications
- ^{67}Cu ($T_{1/2}=61.83$ h) for Therapy and SPECT applications
- ^{89}Zr ($T_{1/2}=78.41$ h) for PET applications

Projects:

- Production and evaluation of Terbium isotopes for therapy and diagnosis.
- Production of ^{44}Ti and development of $^{44}\text{Ti}/^{44}\text{Sc}$ radionuclide generator.
- Production and evaluation of diagnostic ^{44}Sc and therapeutic ^{47}Sc .

	Dy 160 2.329	Dy 161 18.889	
	Tb 159 100	Tb 160 72.3 d	Tb 161 6.91 d β^-
	α 23.2	α 670	
Gd 157 15.65	Gd 158 24.8	Gd 159 18.48 h	Gd 160 21.86
α 254000	α 2.5	β^-	α 1.5
			Gd 161 3.66 min β^-

	Ti 44 60.4 a	Ti 45 3.08 h	Ti 46 8.25	Ti 47 7.44	Ti 48 73.72	Ti 49 5.41	Ti 50 5.18	
	β^+ 78.66... α 1.1 α 0.2	β^+ 1.0 α 720...	α 0.6	α 1.8	α 7.9	α 1.9	α 0.179	
	Sc 43 3.89 h	Sc 44 3.92 h	Sc 45 100	Sc 46 18.7 s	Sc 47 3.35 d	Sc 48 43.67 h	Sc 49 57.2 m	
	β^+ 1.2... α 373...	β^+ 1.2... α 120...	α 10 + 17	α 142	β^+ 0.4... α 159	β^+ 0.7... α 984, 1912, 1038...	β^+ 2.0... α 1782, 1620	
Ca 40 95.941	Ca 41 1.03 · 10 ⁵ s	Ca 42 0.647	Ca 43 0.13s	Ca 44 2.086	Ca 45 163 d	Ca 46 0.004	Ca 47 4.54 d	Ca 48 0.187
α 0.41 α 0.30010	α 0.41 α 0.30010	α 0.30	α 4	α 0.4	β^+ 0.4... α 15	α 0.79	β^+ 0.7... α 1207, 975, 485...	α 1.0

- ❑ M. Ammann, “Using ^{13}N as tracer in heterogeneous atmospheric chemistry”, *Radiochimia Acta* **89**, 831-838 (2001)
- ❑ Home page of the “Laboratory for Radiochemistry and Environmental Chemistry” at PSI: <http://lch.web.psi.ch>
- ❑ Home page of the “Center for Radiopharmaceutical Sciences” at PSI: <http://zrw.web.psi.ch>
- ❑ Home page of the “Laboratory for Energy Systems Analysis” at PSI: <http://lea.web.psi.ch>
- ❑ “Energiespiegel / Mirror On Energy / Le Point Sur L'Energie” at PSI: <http://gabe.web.psi.ch/energie-spiegel/>