The Abdus Salam International Centre for Theoretical Physics

2339-6

Workshop on Atmospheric Deposition: Processesand Environmental Impacts

21 - 25 May 2012

Land-Atmosphere Exchange of Trace Gases and Particles; Principles, Measurement and interpretation of fluxes

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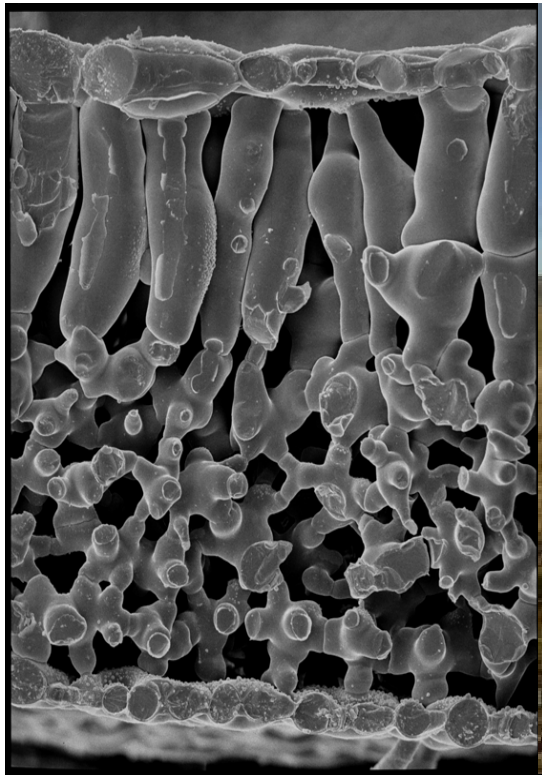
Content

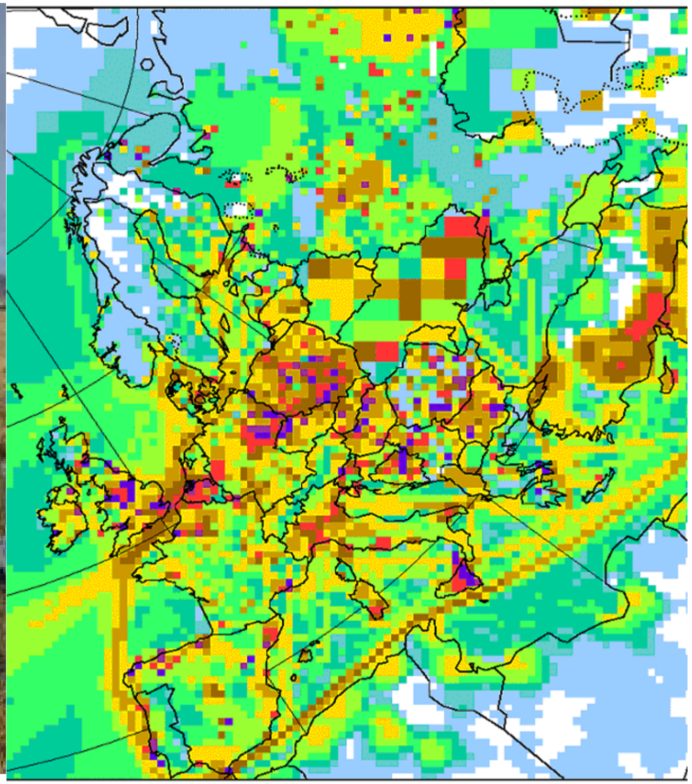
- **Introduction, the role of ecosystems in atmospheric composition**
- **Principles, the processes involved in surface-atmosphere exchange**
- **Resistances, deposition velocities and their components**
- **Measuring surface-atmosphere fluxes**
- **Ozone deposition to natural vegetation**
- **Sulphur deposition**
- **Surface-atmosphere exchange of ammonia (bi-directional exchange)**
- **Particle emission and deposition**
- **Methane and nitrous oxide exchange**

•Introduction, the role of ecosystems in atmospheric composition

- The earth is unique in having an atmosphere largely of biological origin
- For most trace gases exchange is regulated at the surface-atmosphere interface
- The policy needs have set priorities for research (SO_2, NO_2, O_3…..)
- Surface –atmosphere exchange is a key source or sink of gases and particles contributing to Acidification eutrophication, ozone and other photochemical oxidants, climate change

SCALE





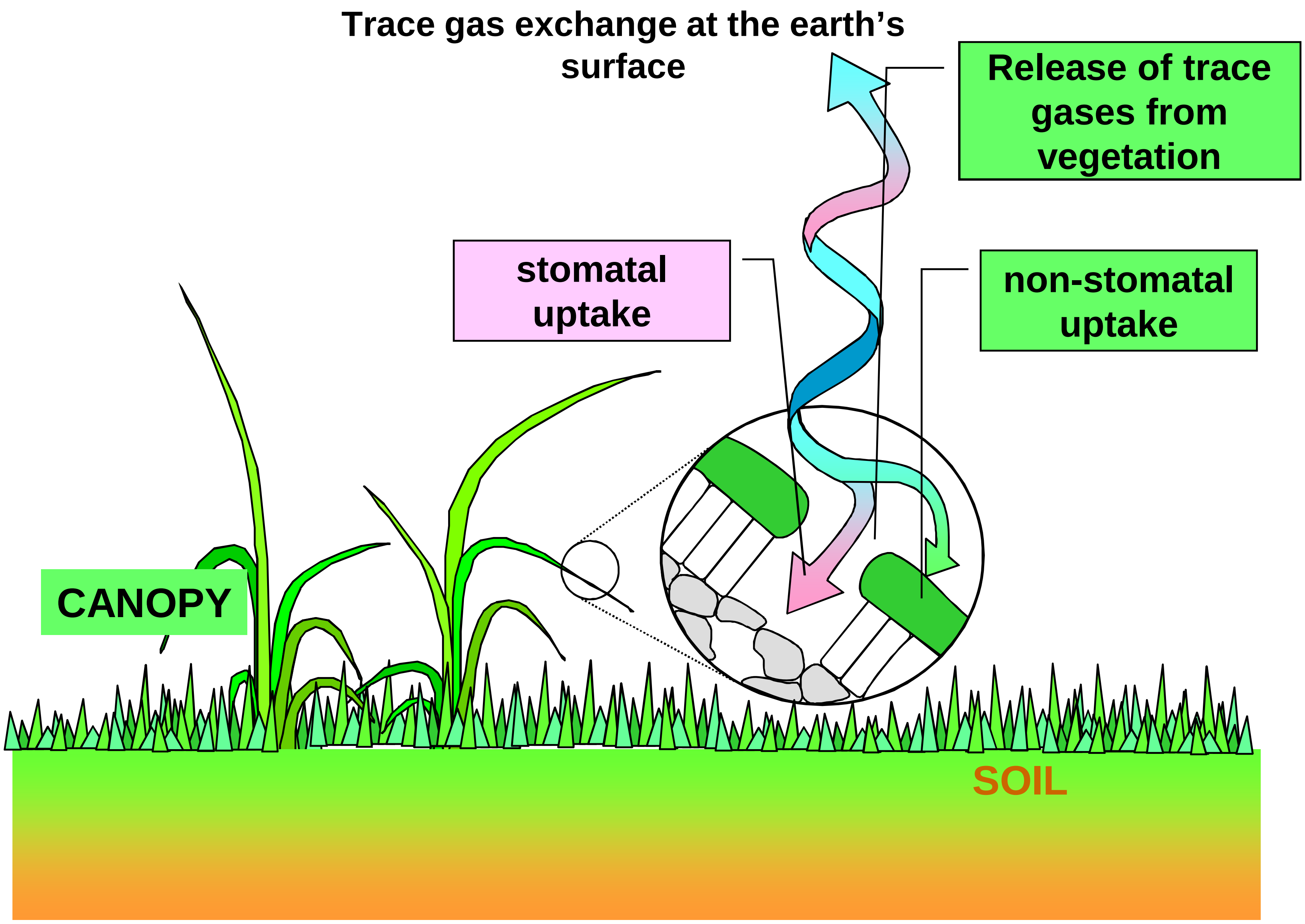
Molecular to um
Controls the exchange

Measurement scale
Landscape (100-1000m

Application: region to
continent 100km to10000km

The transfer process

- Turbulent transfer in the free atmosphere
- For trace gases, molecular diffusion becomes important very close to surfaces (<mm)
- At the surface for many of the trace gases chemical interaction with the surface components and water are very important

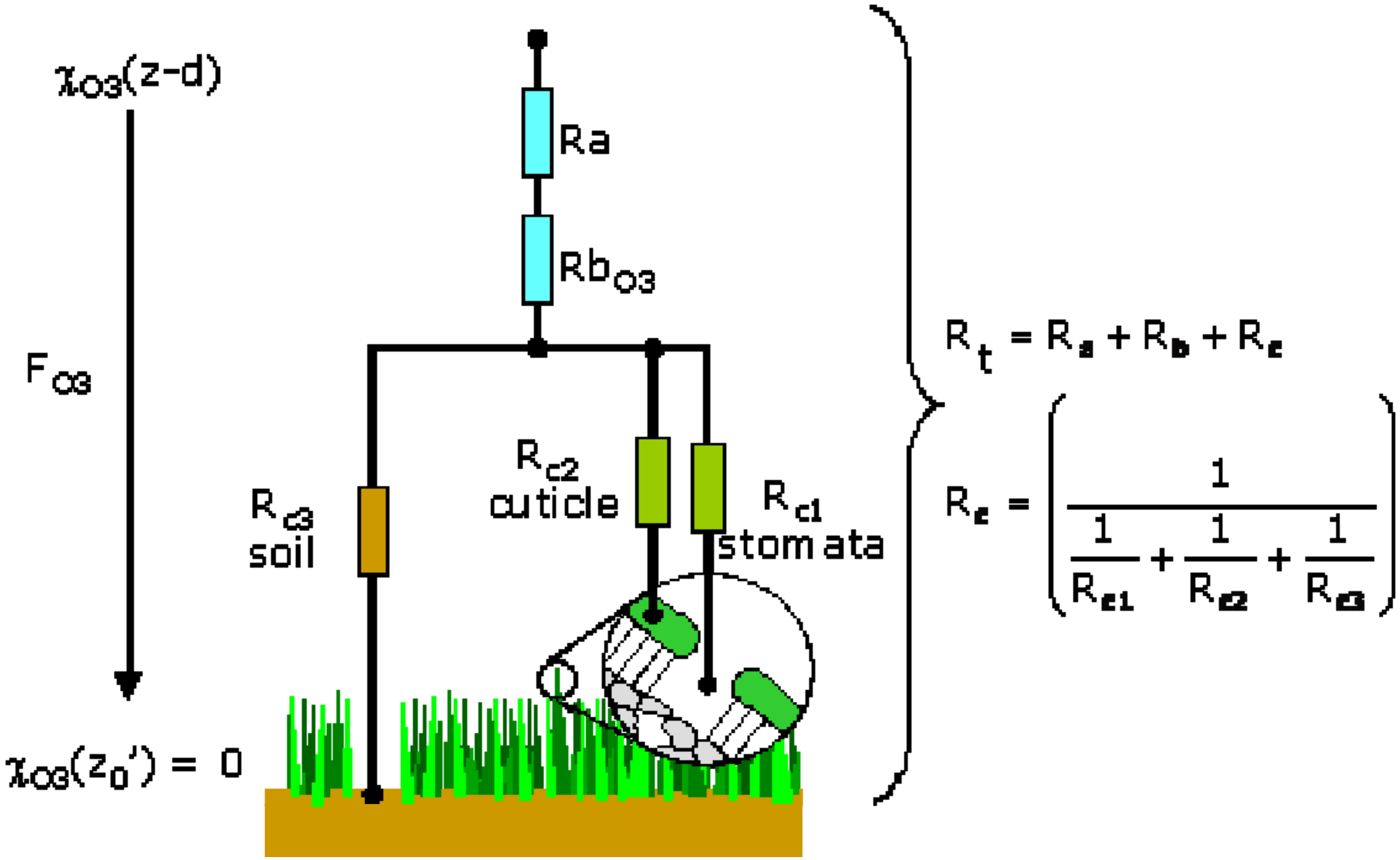


Trace gas exchange at the earth's surface
Release of trace gases from vegetation
stomatal uptake
non-stomatal uptake
CANOPY
SOIL

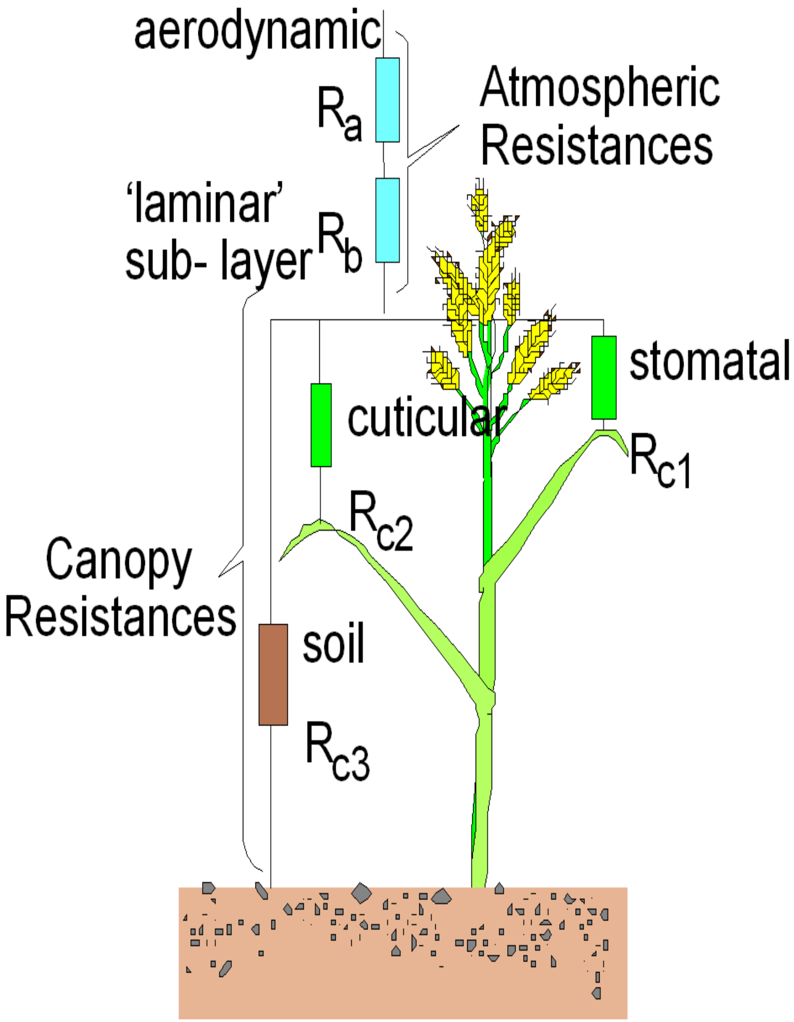
Dry Deposition

Definition: The direct transfer of pollutants to terrestrial surfaces

Simple resistance models



The resistance analogy



$$v_d = \frac{\text{flux}}{\text{concentration}} = \frac{1}{R_t}$$

$$R_t = R_a + R_b + (1/R_{c1} + 1/R_{c2} + 1/R_{c3})^{-1}$$

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ATMOSPHERIC ENVIRONMENT

sition interactions

Review

Atmospheric composition change: Ecosystems–Atmosphere interactions

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Flux measurement techniques

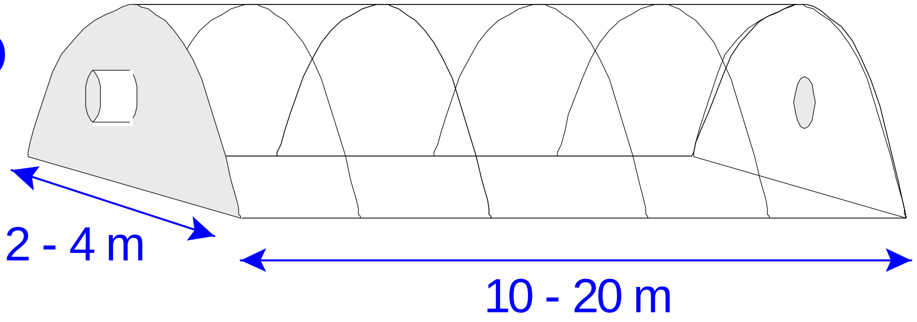
- Chamber methods   plot scale
- Micrometeorology….(field to landscape)
- Boundary layer budget (landscape to region)
- Remote sensing (global)

② overcomes small scale variability

② provides substantial spatial averaging

Mega-chamber N_2O

20 - 80 m^2



CLIMEX -
catchment “greehhouse”
study NO, NO_2, (CH_3, CO_2, O_3)

450 m^2

Chamber Effects

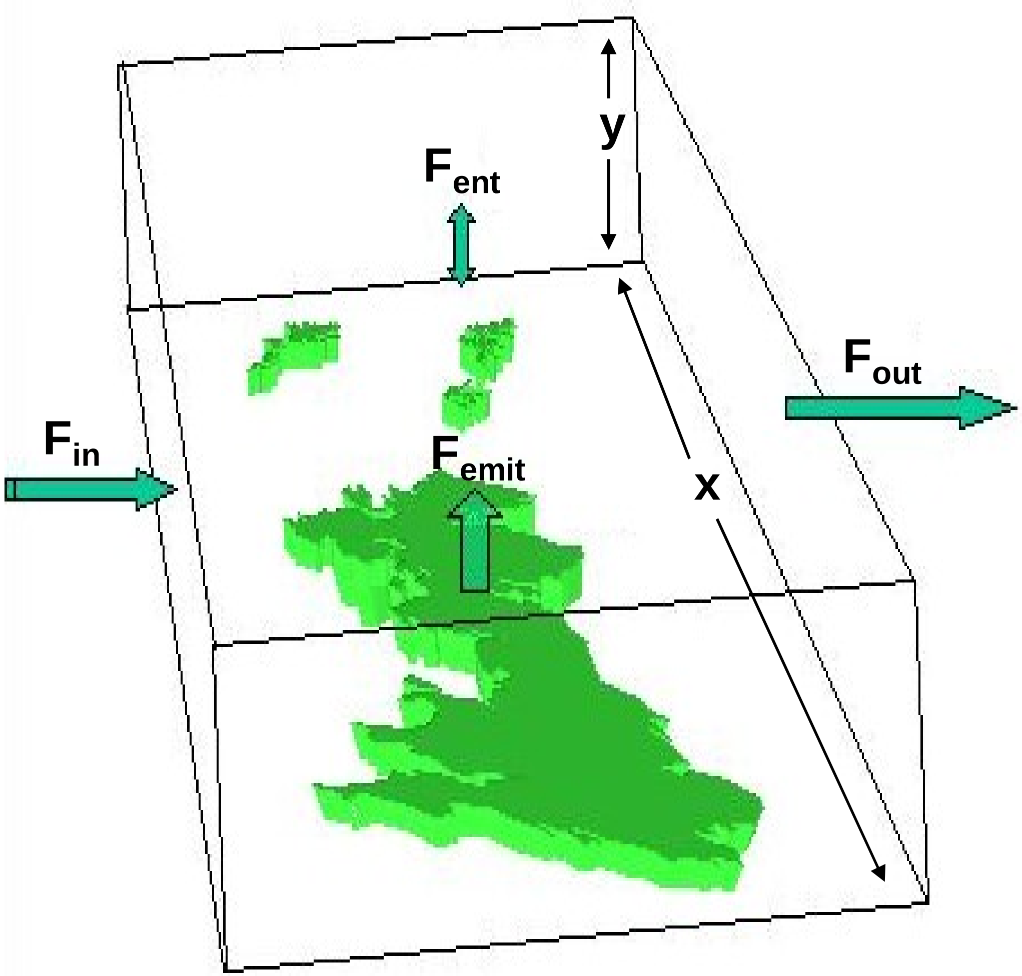
1. Δtemperature +10 ^{o}C over 30 minutes

   CH4 emission varies with temperature but the chamber temperature and water temperature do not have the same rseponse time so that long term data cannot be used to quantify this effect (< 10 % ?)

2. Δpressure

   A very small effect as there is a large hole in the chamber wall to maintain pressure.

Estimating Annual Emissions – Box Technique



- $F_{emit} = F_{out} – F_{in} – F_{ent}$
- $F_{in} = \iint u_{in}(y,z)c(y,z)dydz$
- $F_{out} = \iint u_{out}(y,z)c(y,z)dydz$
- $F_{ent = 0}$

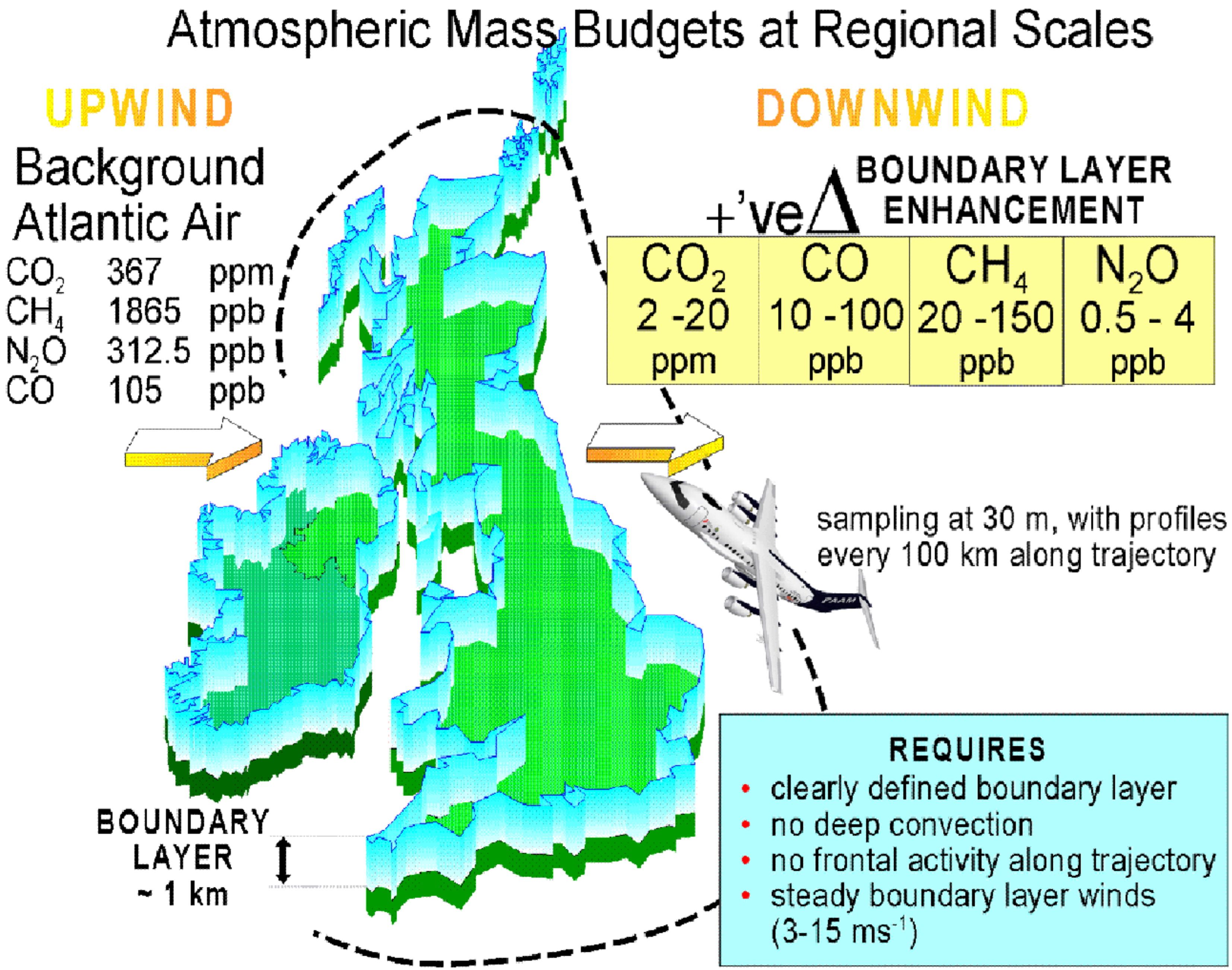
F_{emit} = Flux emitted

F_{in} = Flux in

F_{out} = Flux out

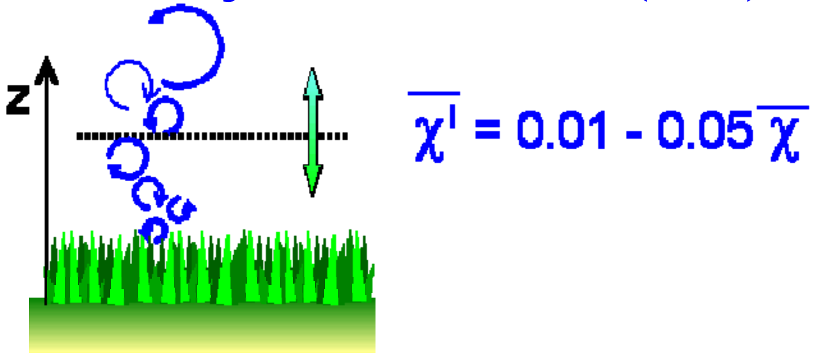
u = wind speed

c = concentration


Atmospheric Mass Budgets at Regional Scales
UPWIND
Background Atlantic Air
CO_2 367 ppm
CH_4 1865 ppb
N_2O 312.5 ppb
CO 105 ppb
DOWNWIND
+'ve Δ BOUNDARY LAYER ENHANCEMENT
CO_2 2 -20 ppm
CO 10 -100 ppb
CH_4 20 -150 ppb
N_2O 0.5 - 4 ppb
sampling at 30 m, with profiles every 100 km along trajectory
BOUNDARY LAYER ~ 1 km
REQUIRES
• clearly defined boundary layer
• no deep convection
• no frontal activity along trajectory
• steady boundary layer winds ($3\text{-}15\ ms^{-1}$)

Micrometeorological Methods

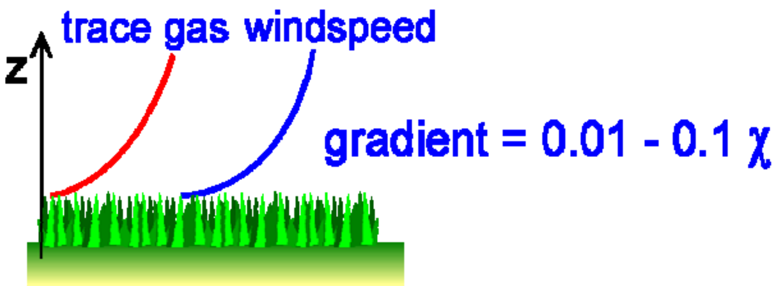
Eddy-covariance (EC)



$$\mathrm{flux}_\chi = \overline{w'\chi'}$$

w' - fluctuation in vertical wind speed
χ' - deviation from mean concentration

Aerodynamic flux-gradient



$$\mathrm{flux}_\chi = K_\chi \frac{\partial \chi}{\partial z}$$

with stability correction:

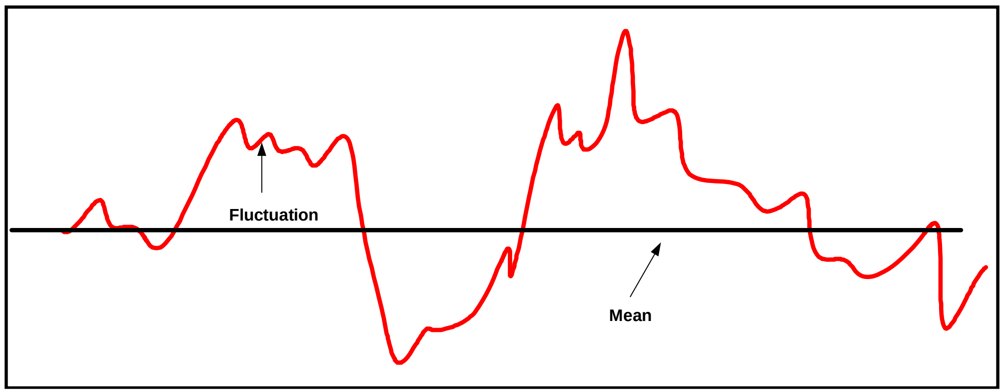
$$\mathrm{flux}_\chi = k u_* \frac{\partial \chi}{\partial [\ln(z-d) - \Psi_H\{\varsigma\}]}$$

Eddy Covariance

- Pros
  - Direct , not inferred (e.g. K-theory), measure of the flux density between the atmosphere and biosphere
  - *In situ*
  - Quasi-continuous
  - Integrative of broad area
  - Introduces no artifacts, like chambers
- Con
  - Expensive
  - Suffers from errors in complex terrain or advective conditions
  - Modern chemical sensors require electrical power, limiting where the method can be employed
  - Not amenable for all trace gases, if fast sensor is not available

Eddy Covariance Technique

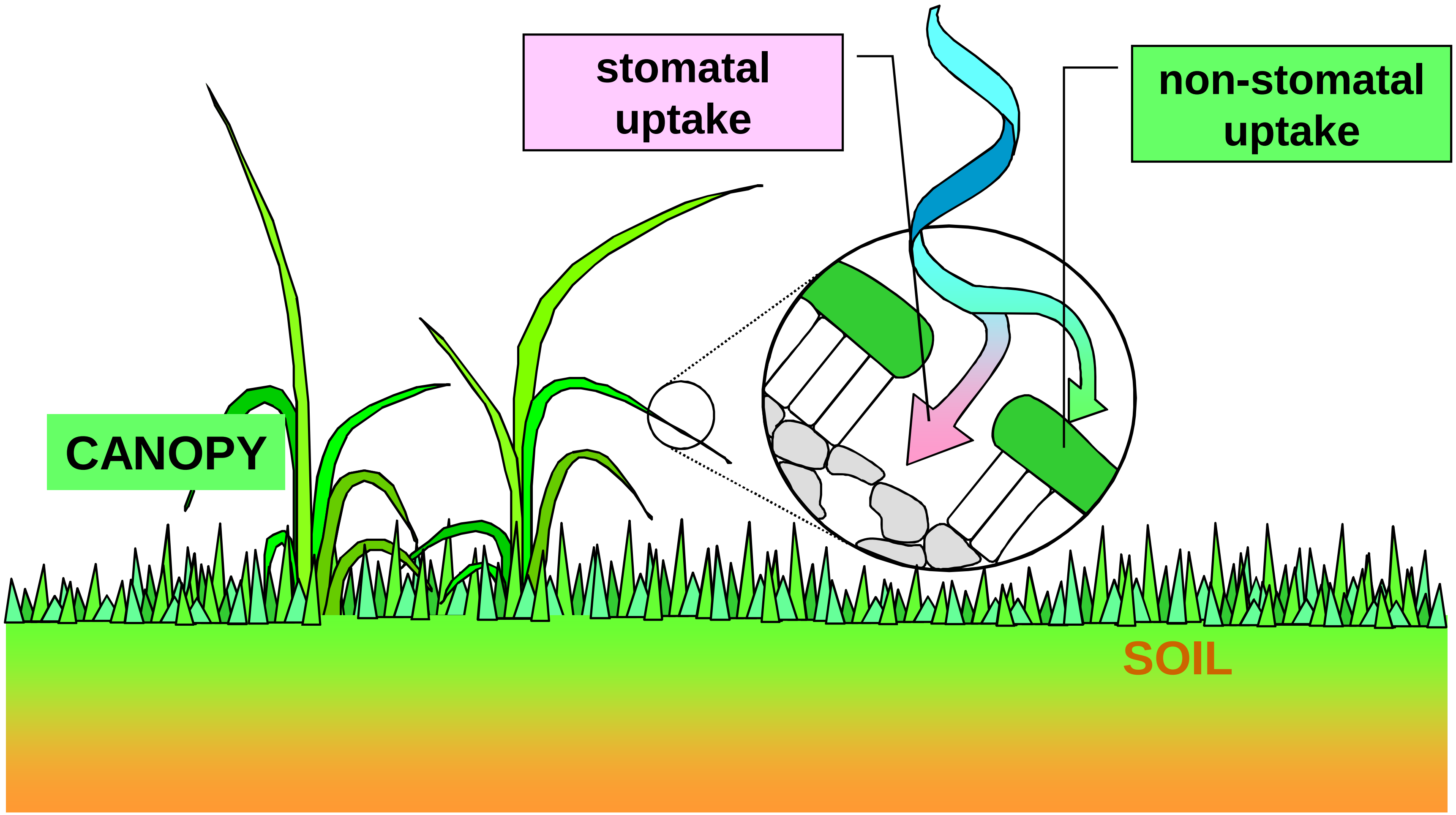
$$F = \overline{\rho w s} \sim \overline{\rho_a} \cdot \overline{w' s'} \qquad s = (\frac{\rho_c}{\rho_a})$$



Ozone

- Identifying the different sinks for ozone at the surface
- Separating stomatal and non-stomatal fluxes to vegetation
- Understanding the effects of soil moisture, leaf temperature and humidity on ozone flux.
- Understanding non-stomatal ozone deposition
- The importance of O_3 deposition to vegetation in episode conditions and feedbacks from hot dry summers (2003).
- Long term measurements of ozone flux

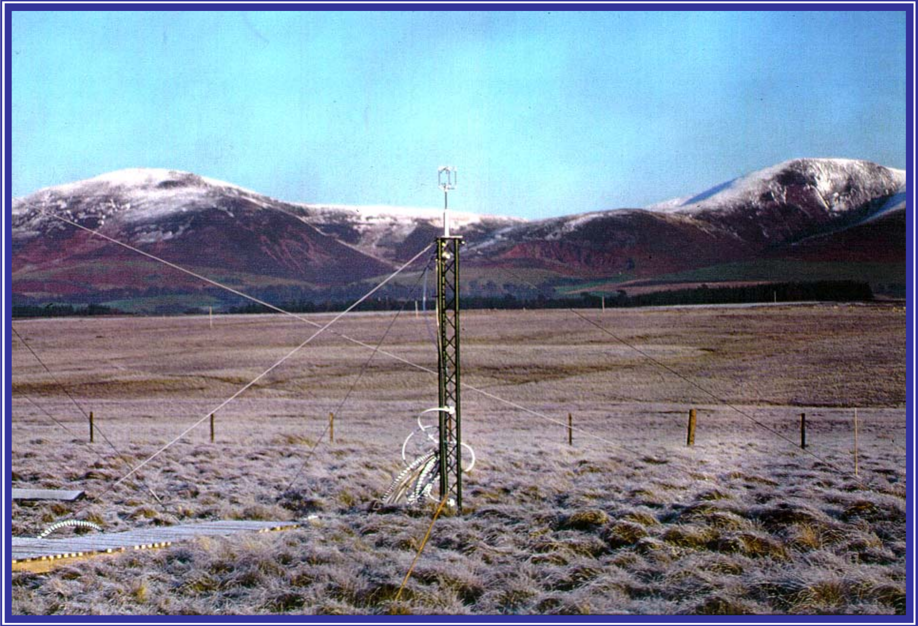
Ozone exchange at the earth's surface



ROFI: A New "Rapid Ozone Flux Instrument"

- Measure O_3 and H_2O fluxes using the eddy-correlation method.
- Low power instruments can be run using a wind turbine and solar panel.
- Allow measurement of total, stomatal and non-stomatal ozone deposition in remote locations without access to mains power.

Auchencorth Moss



55º 47’ 30’’ N, 3º 14’ 20’’ W
Altitude 270 m a.s.l
Ombrotrophic mire, > 1000 ha

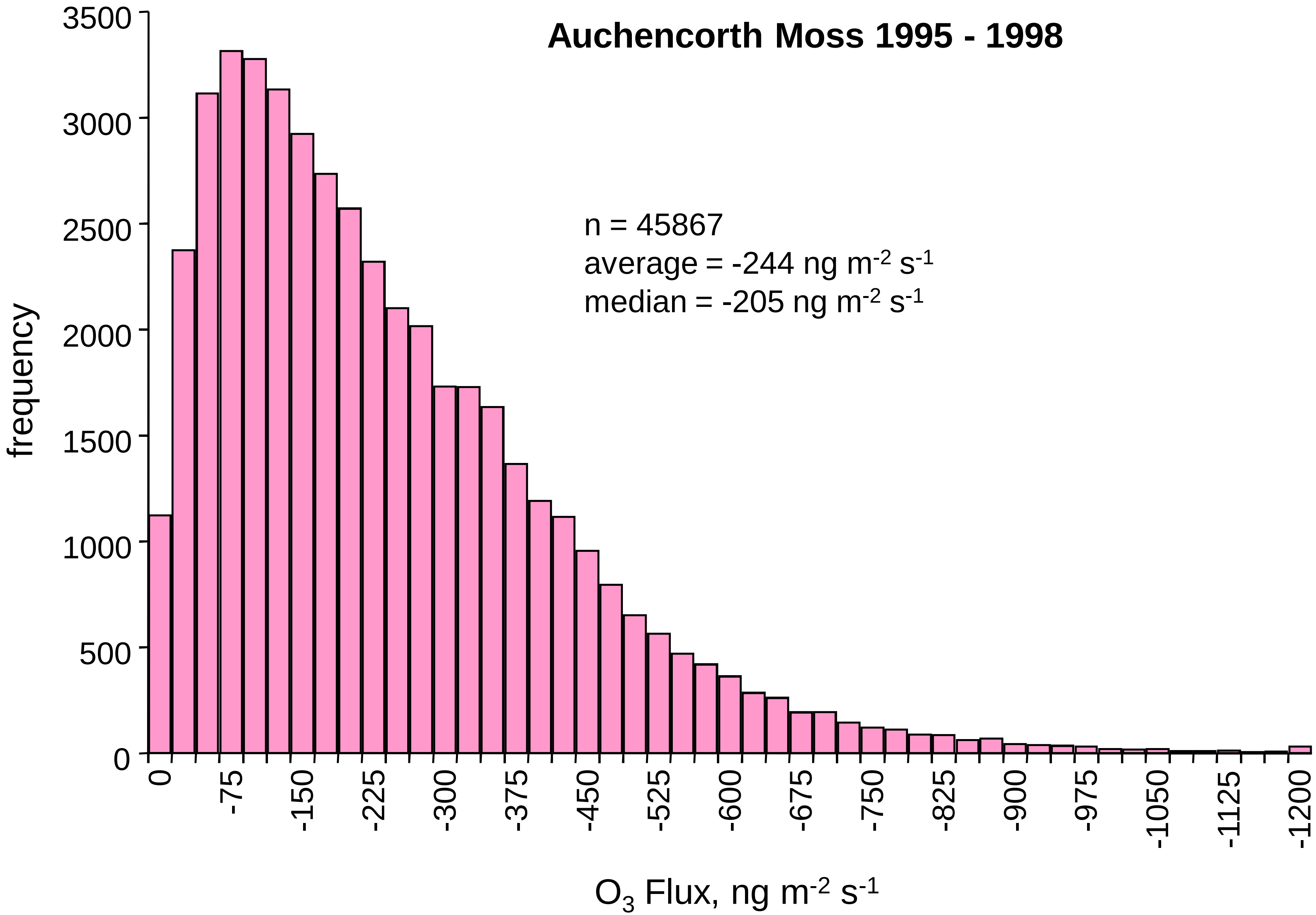
- moorland canopy height 0.2 – 0.4 m
- LAI 4-6 summer, 2-3 winter
- Deschampsia flexuosa, molinia caerulea, eriophorum vaginatum, festuca ovina, calluna vulgaris, vaccinium myrtillus, carex nigra, sphagnum spp.

- ❖ Continuous flux measurements from 1995 to 1998.
- ❖ Eddy-covariance (EC) u_*, C, λE, CO_2
- ❖ EC & flux-gradient O_3 (NO, NO_2, SO_2, NH_3)
- ❖ Bowen ratio λE, C

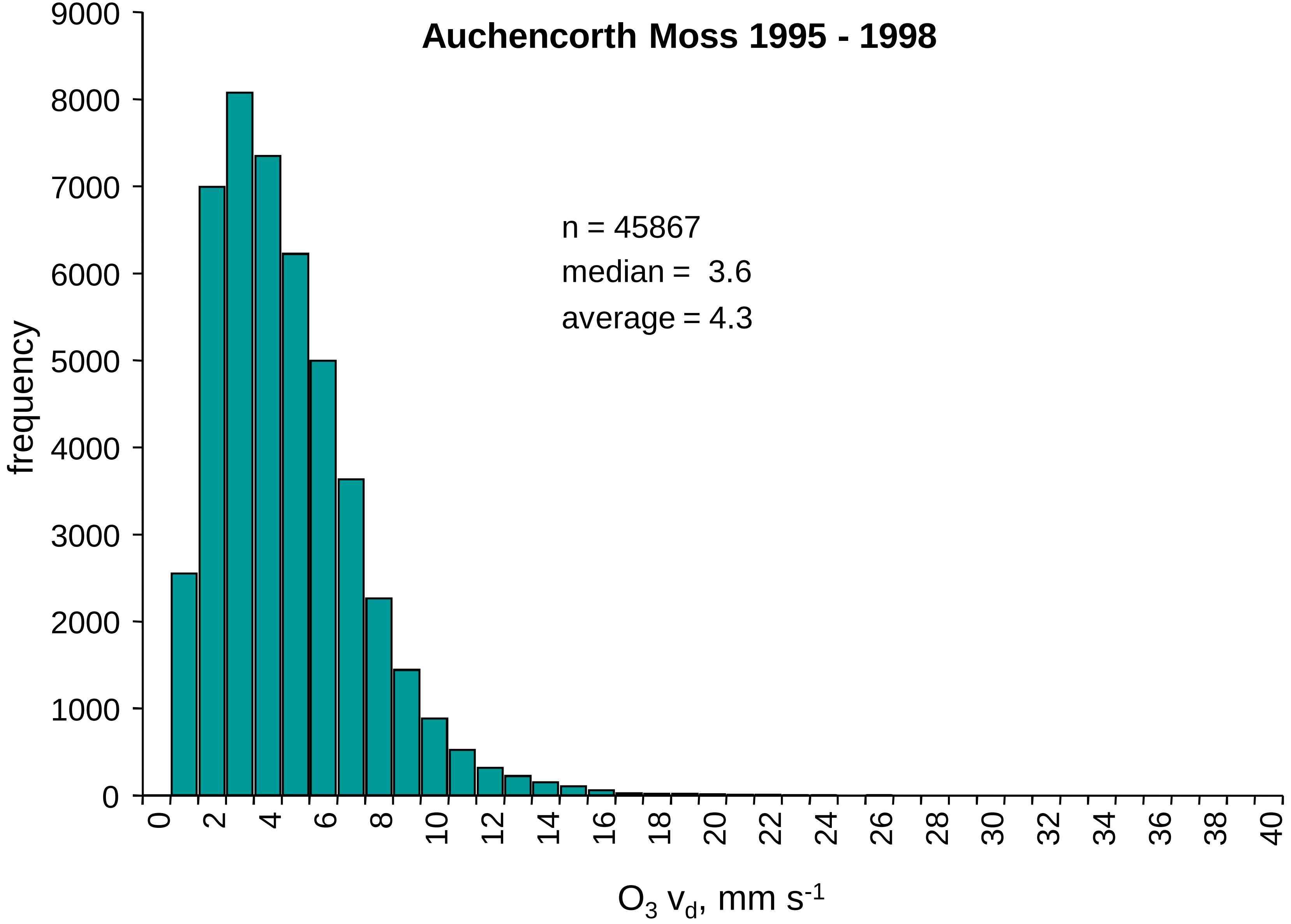
Data capture, ~24000 hours of flux data:

concentration	fluxes
94%	68%

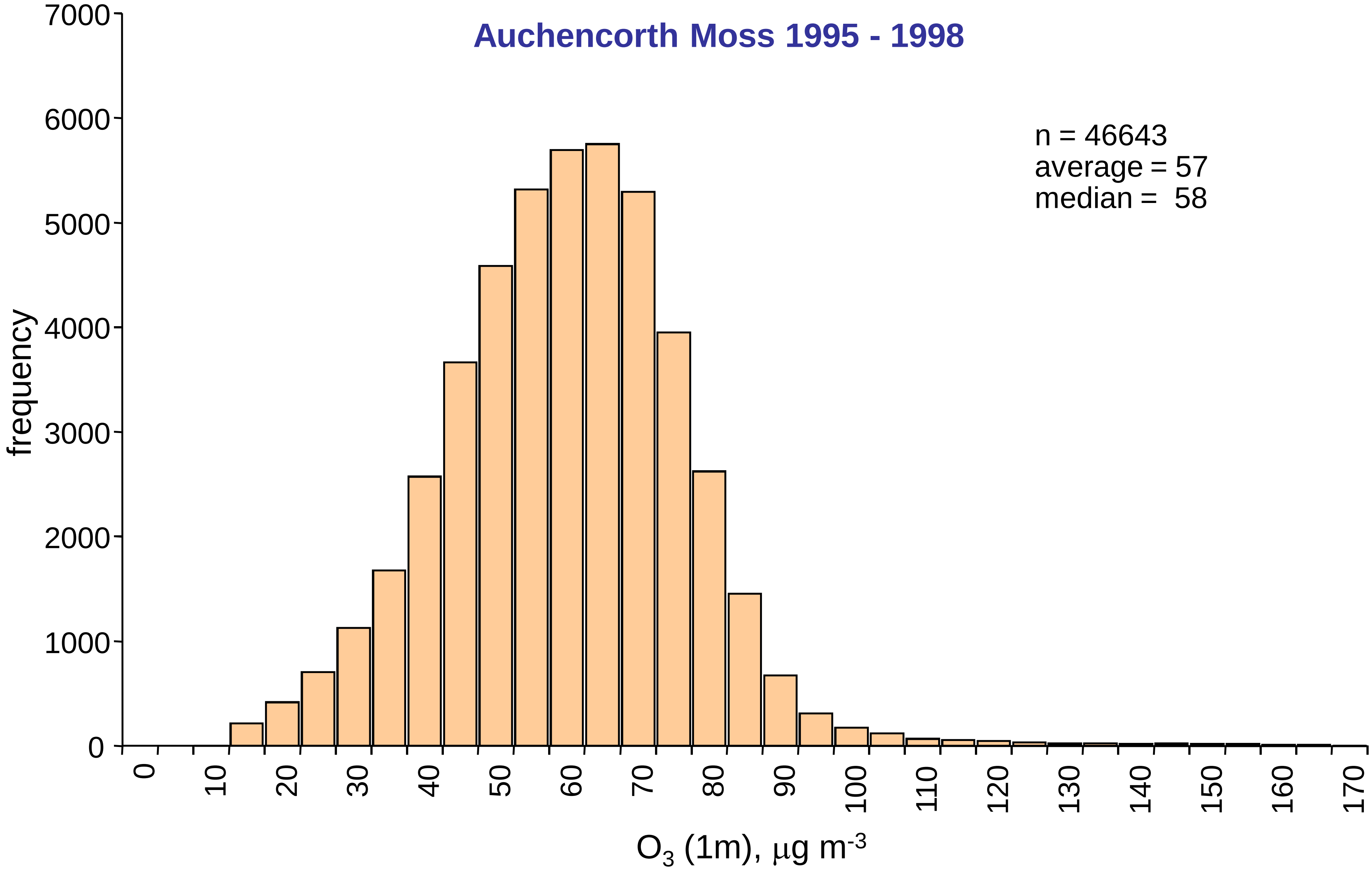
Frequency Distribution: O_3 deposition flux.



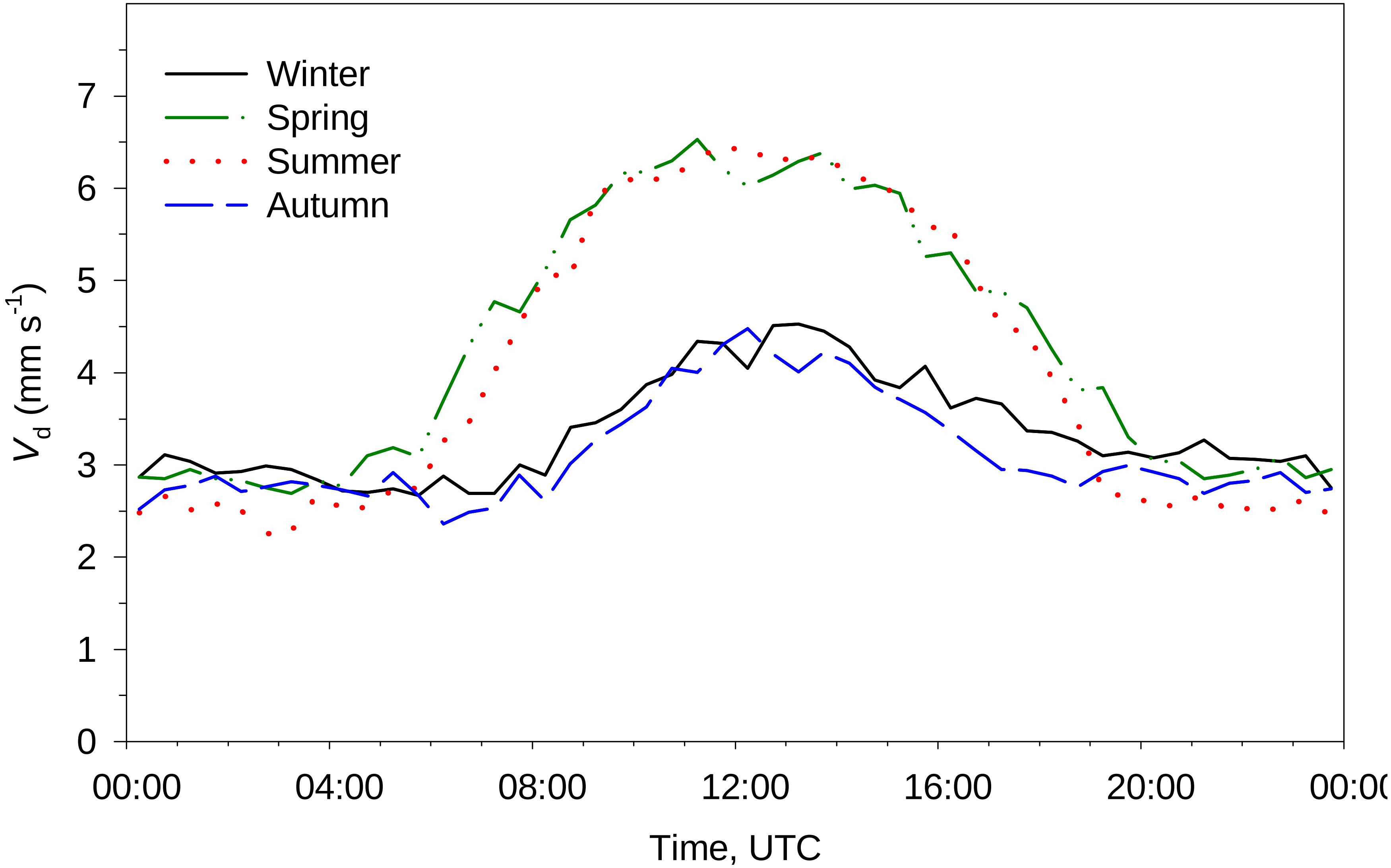
Frequency Distribution: O_3 deposition velocity.



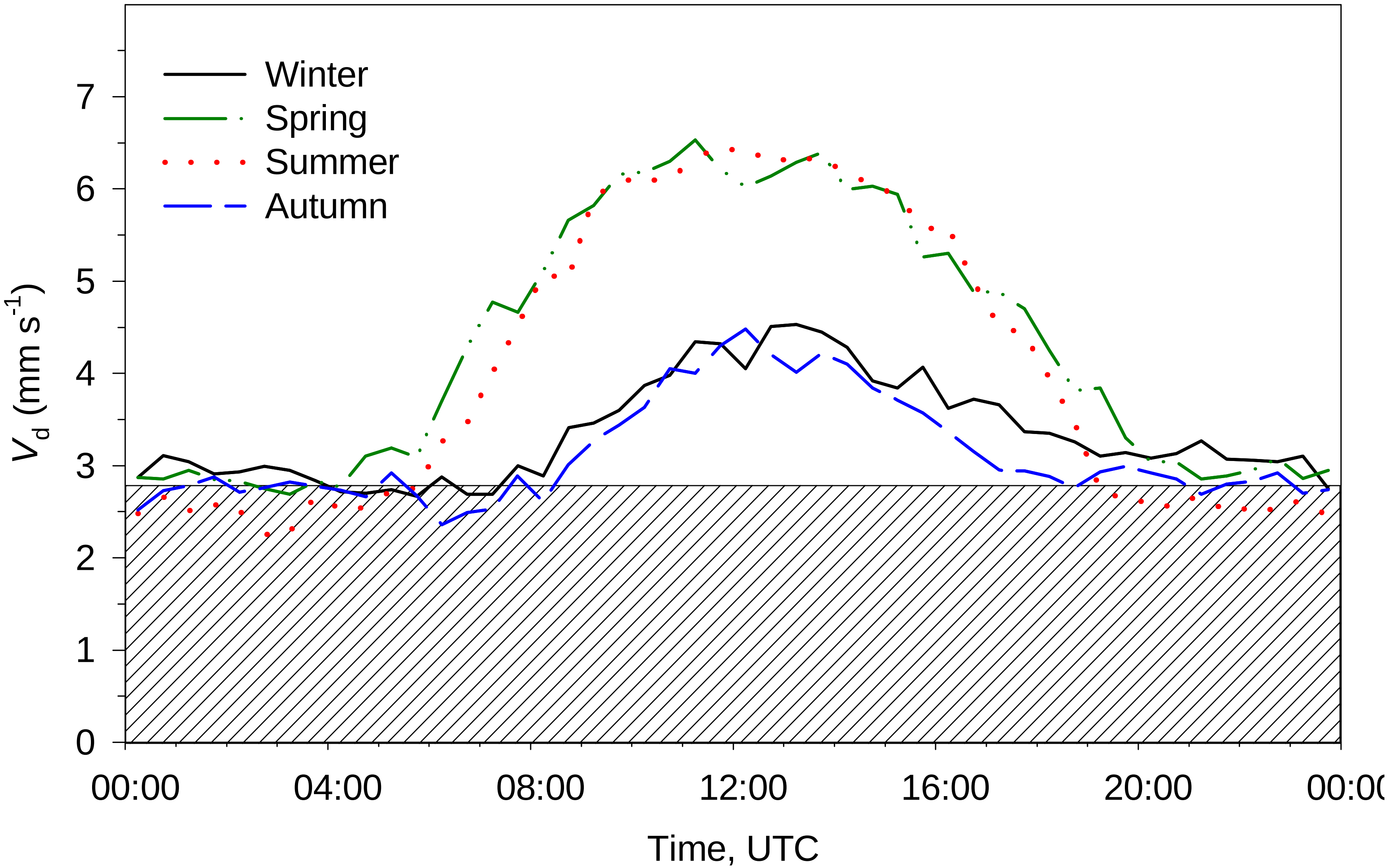
Frequency Distribution: O_3 concentration.



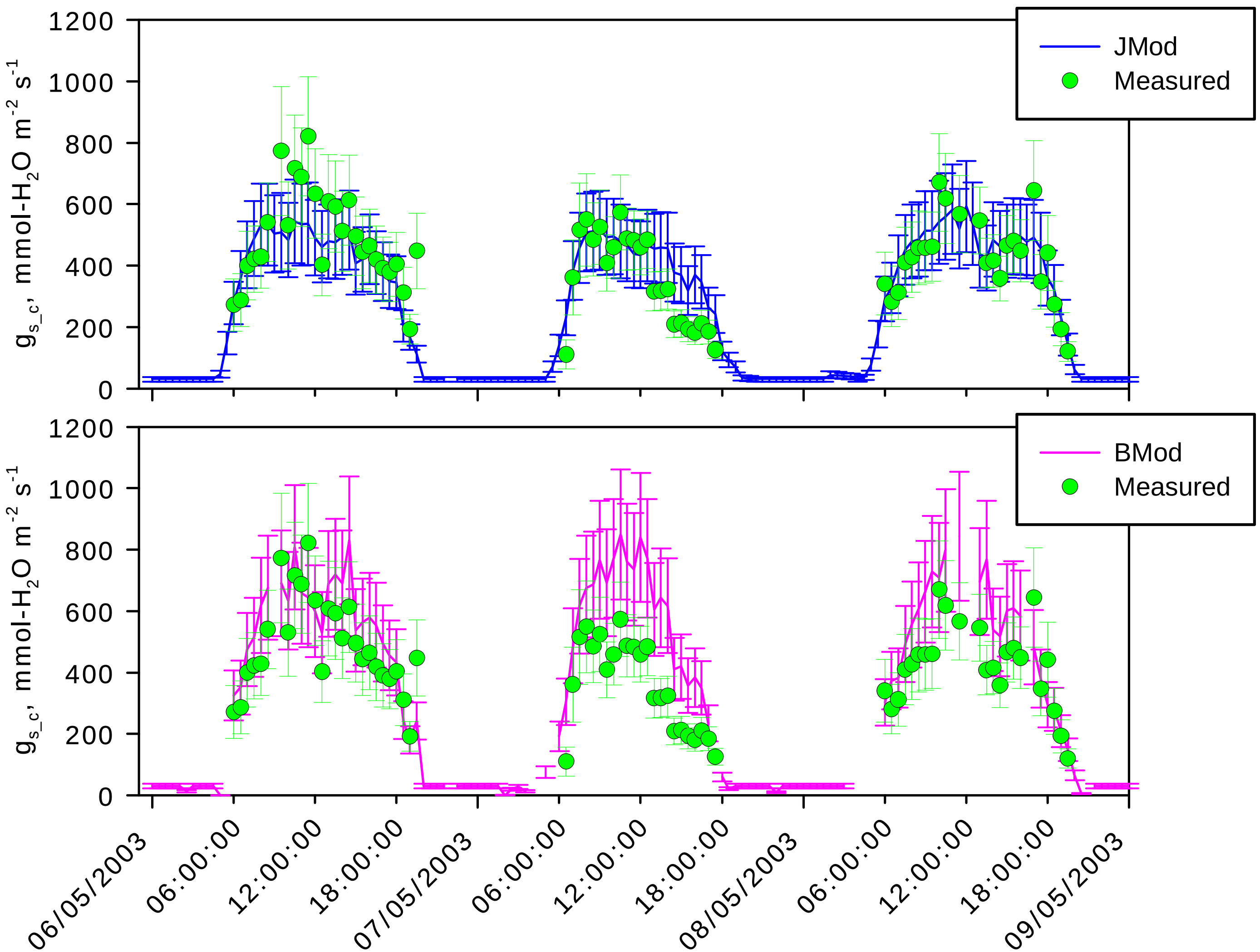
1995 – 98 Seasonal average deposition velocities at Auchencorth Moss



1995 – 98 Seasonal average deposition velocities at Auchencorth Moss

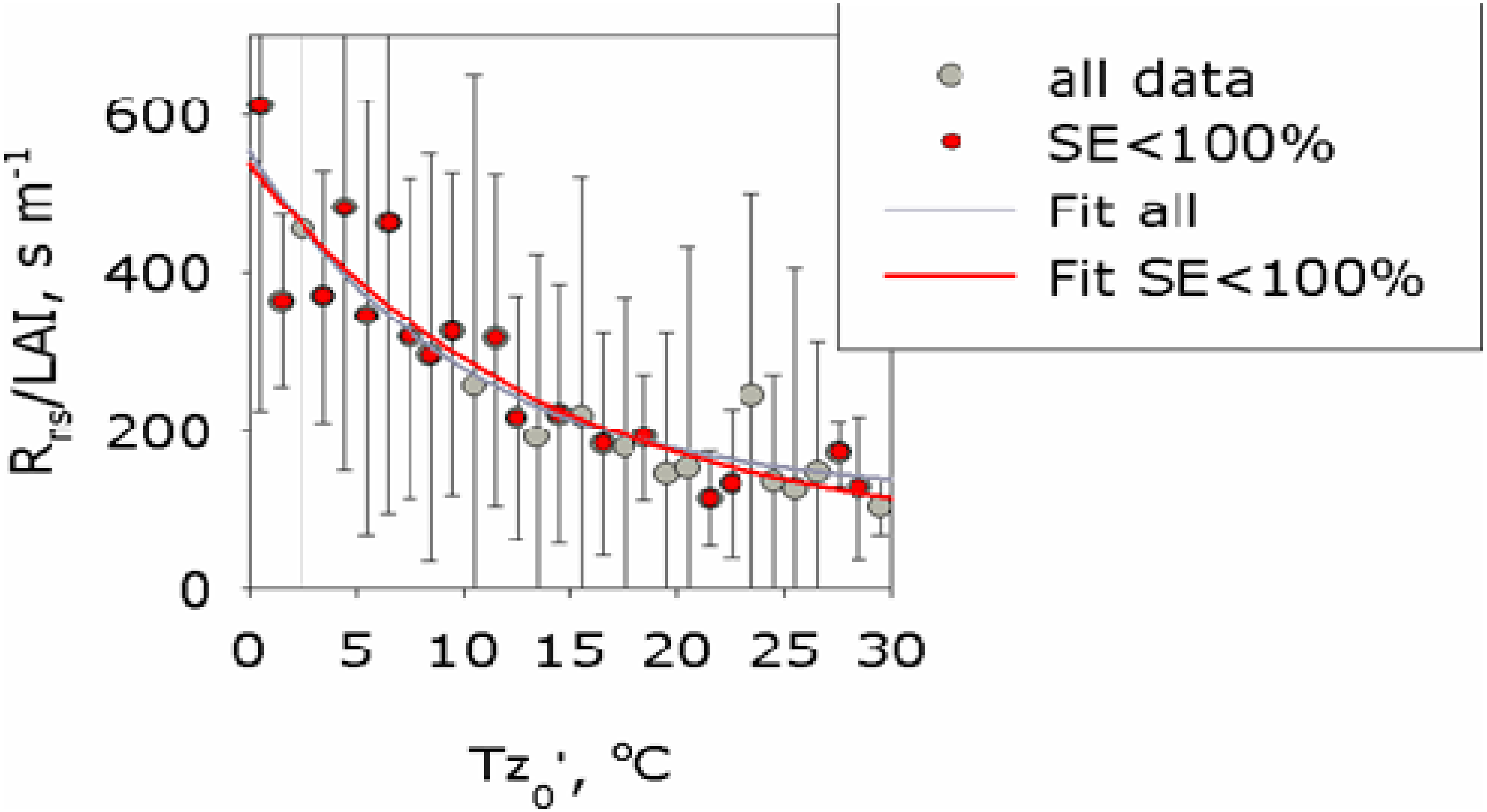


Modelled and measured stomatal fluxes

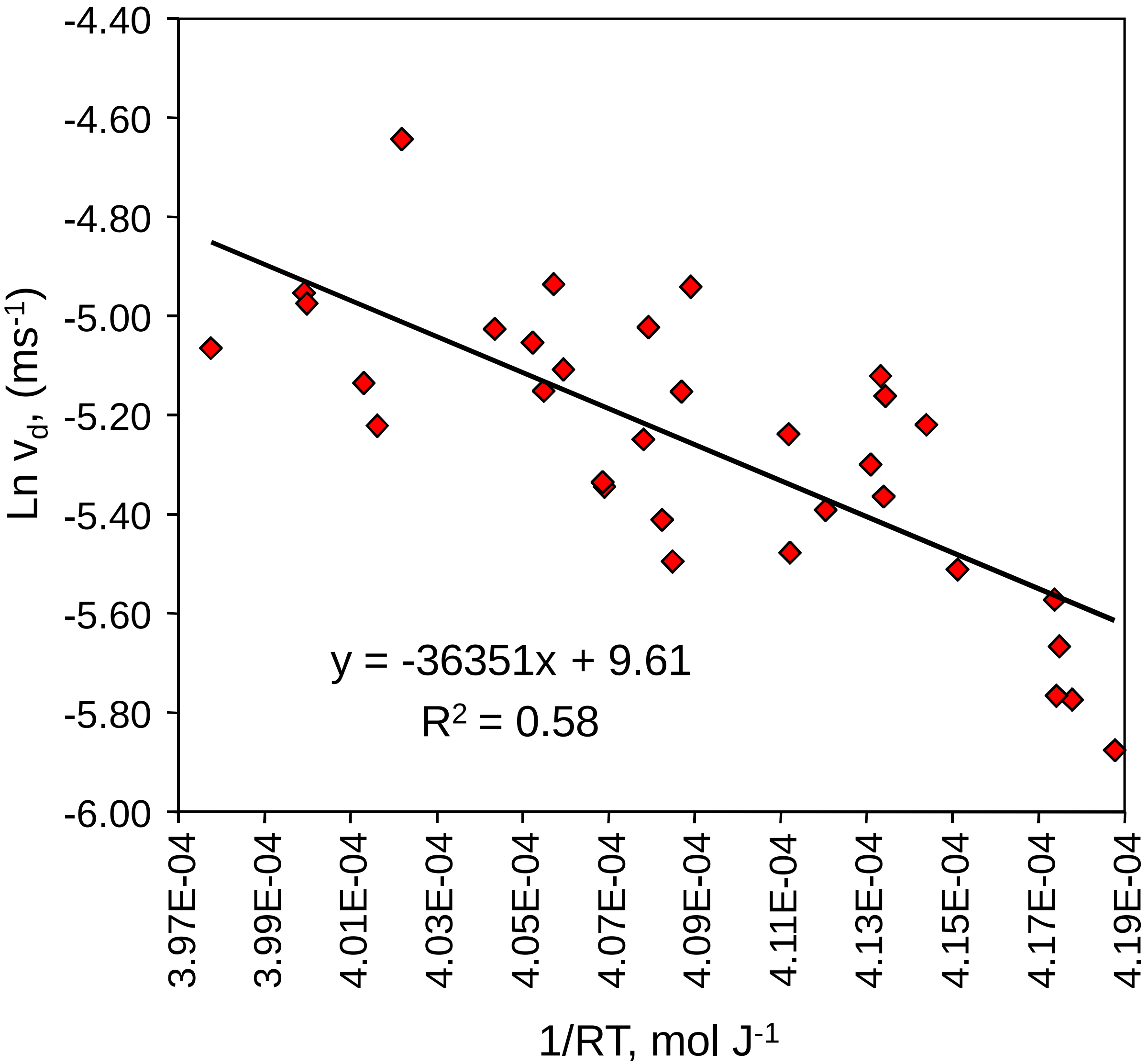


- The mechanisms of ozone depletion on leaf surfaces and within vegetation

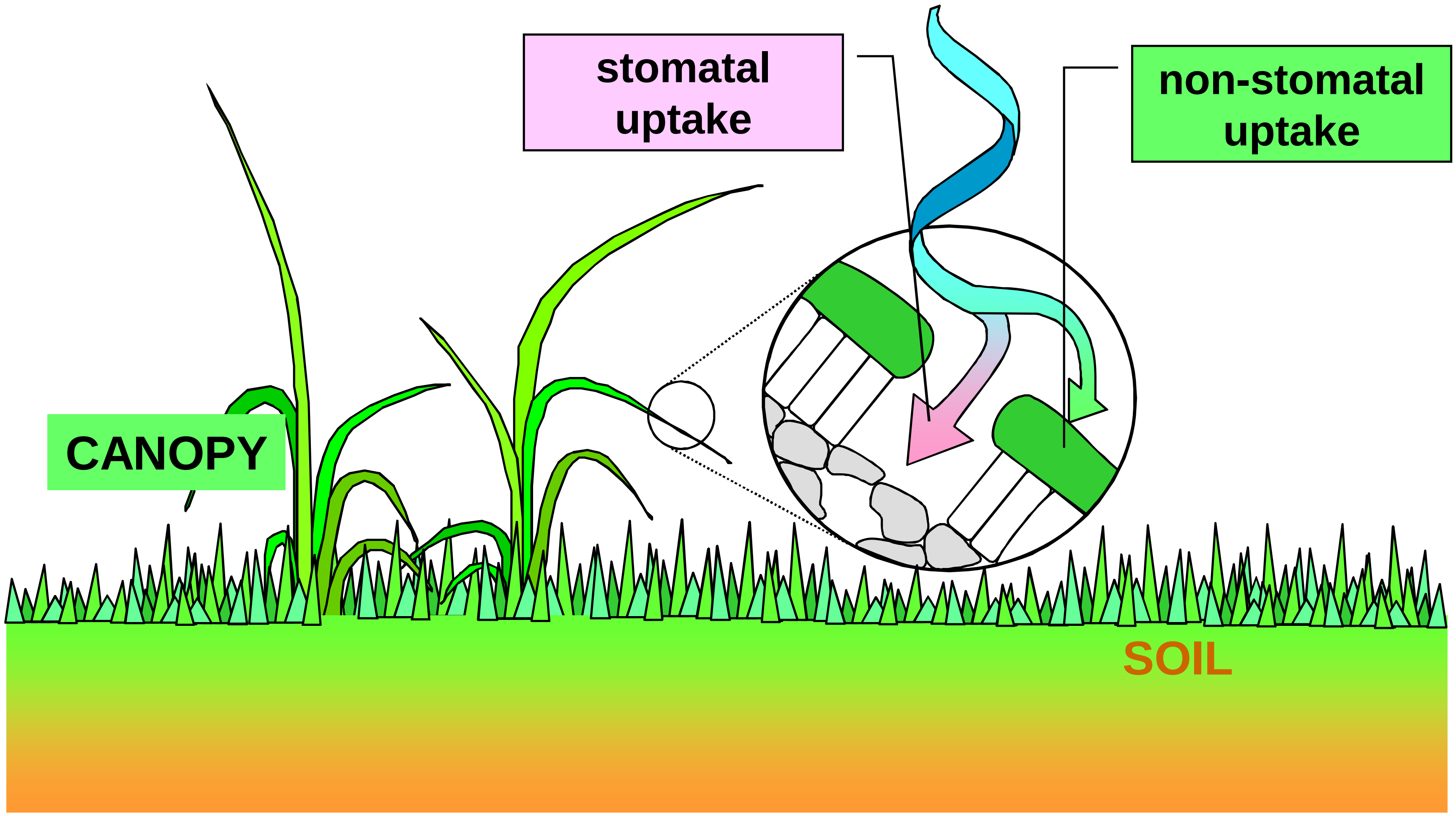
Effects of leaf surface temperature on canopy resistance



Arrhenius plot for the reaction rate of ozone on non-stomatal surfaces



Pathways of Ozone Deposition

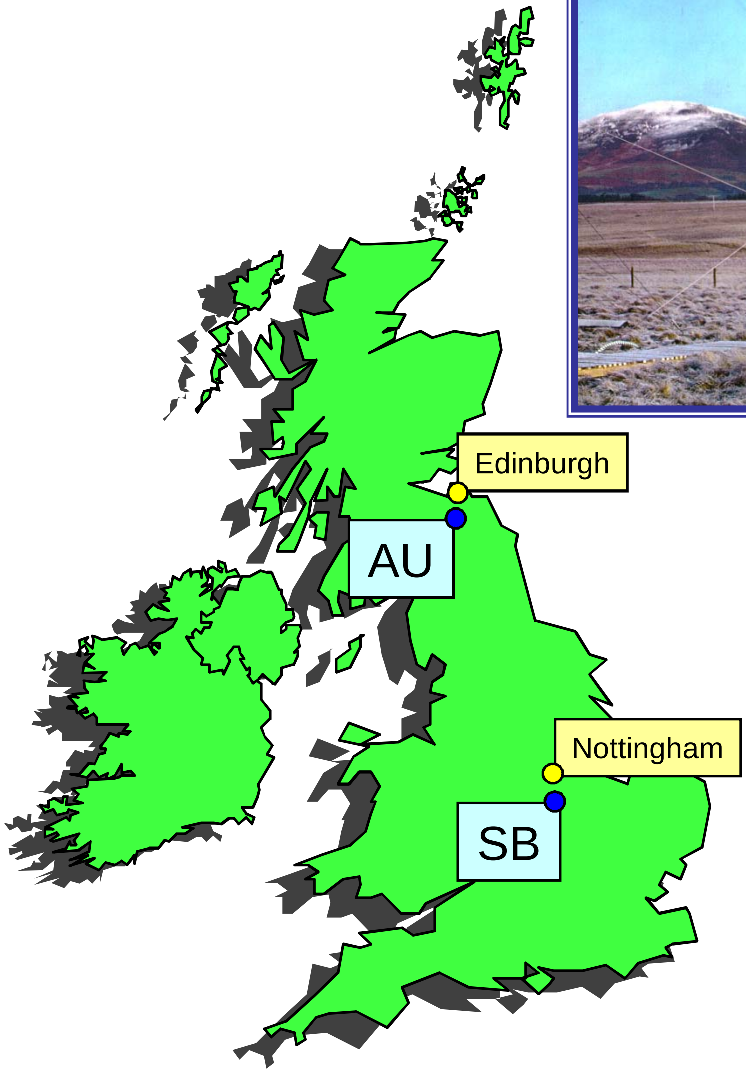


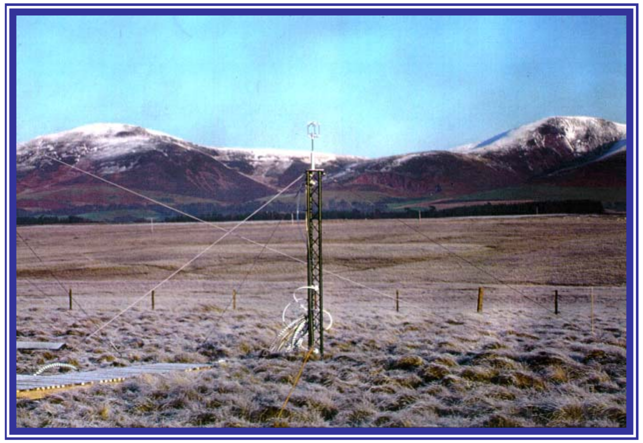
- Stomatal flux cf AOT40
- Ozone risk assessments by the two methods over Europe

Sulphur Dioxide

- Processes regulating exchange at the surface, competition between stomata and leaf surface sinks, the effect of leaf surface wetness and the control of canopy resistance by chemistry in moisture on leaf surfaces.
- Non-linearities and the effects of NH3/SO2 ratios at regional scales.
- Long-term trends in deposition velocity over Europe

Auchencorth Moss (AU) & Sutton Bonnington (SB)

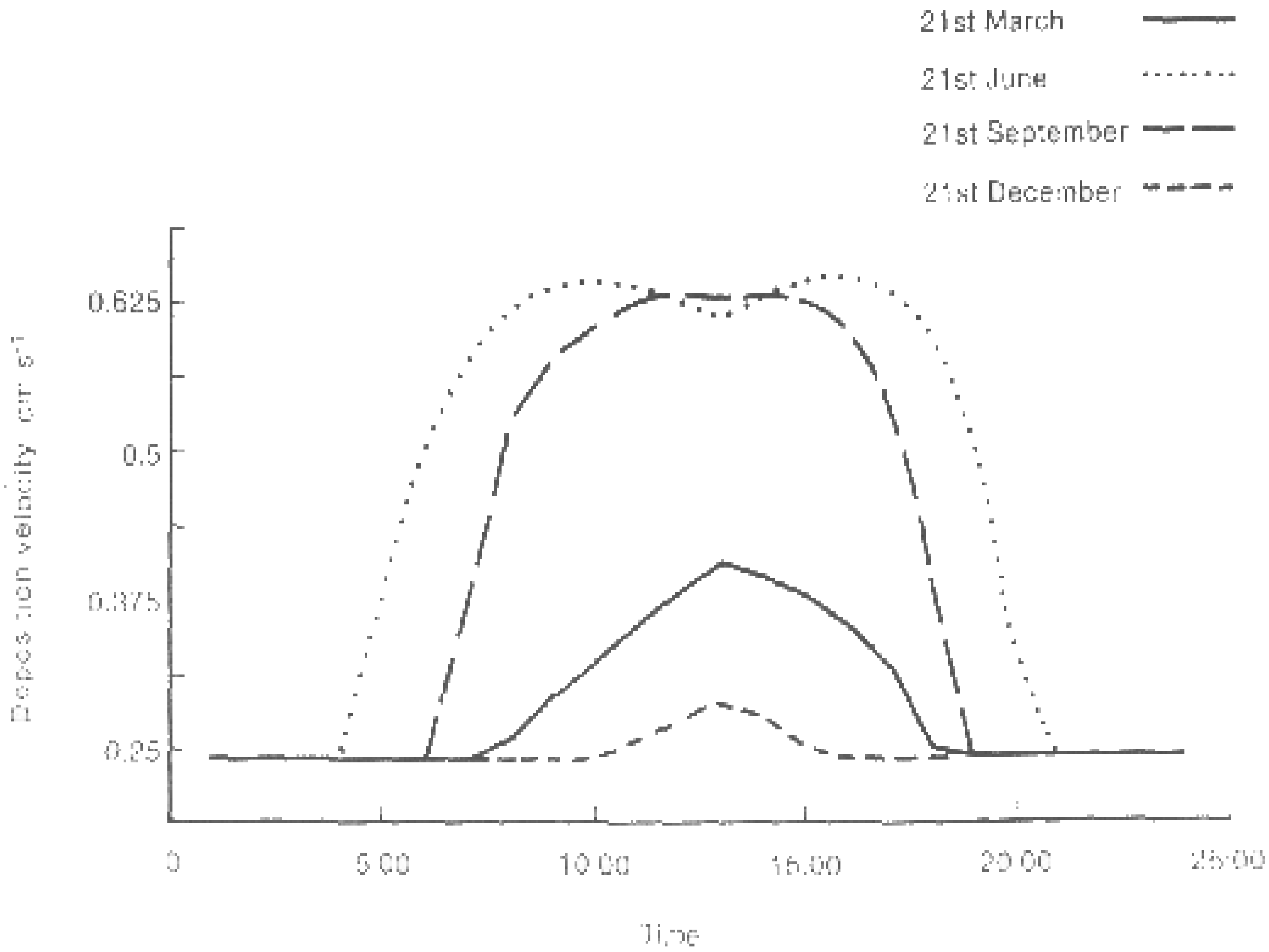




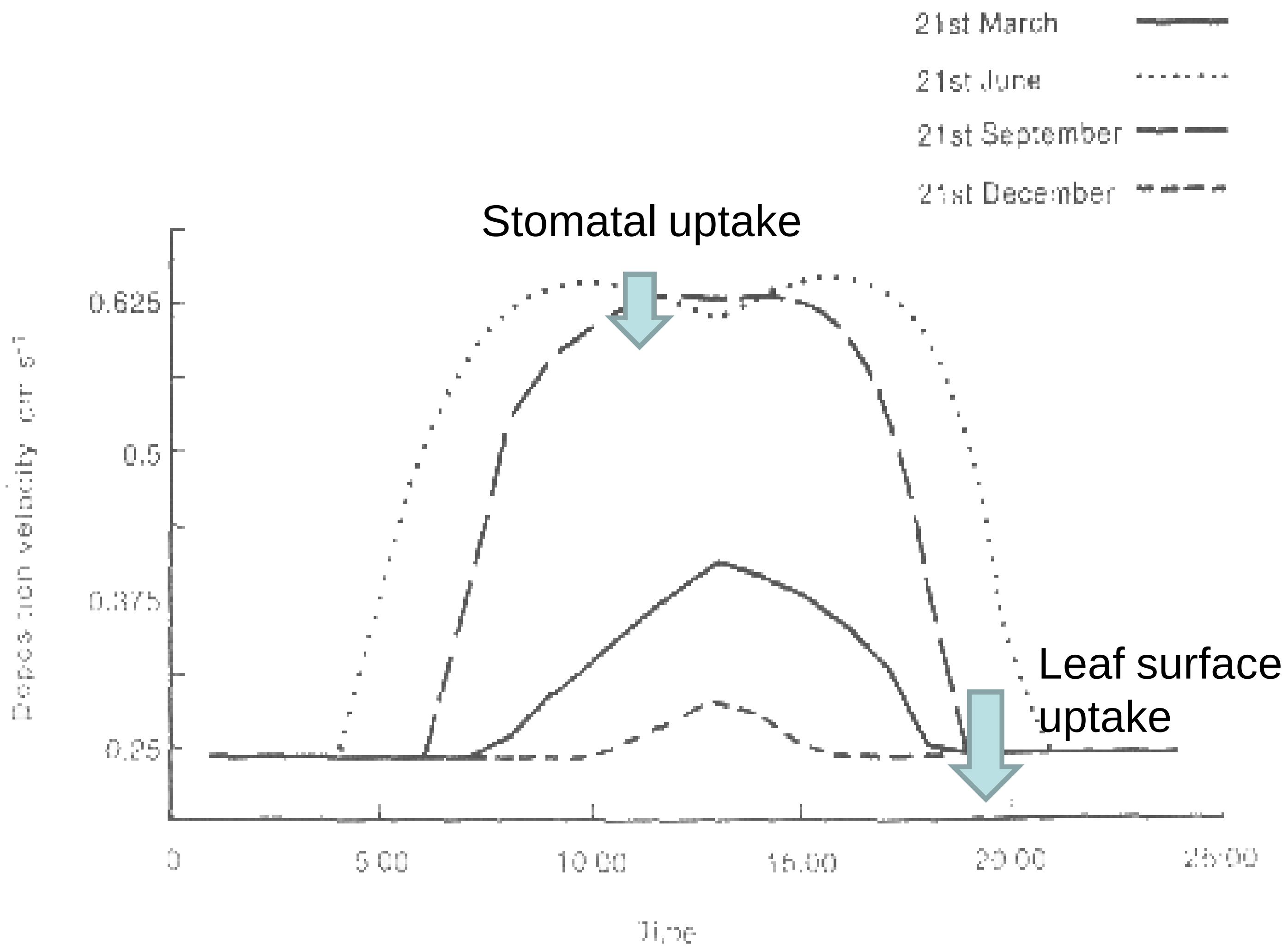
Auchencorth Moss
55º 47’ 30’’ N, 3º 14’ 20’’ W
Altitude 270 m a.s.l
Ombrotrophic mire, > 1000 ha

Sutton Bonnington
52º 50’ 24’’ N, 1º 15’ E
Altitude 45 m a.s.l
Arable farmland, > 4 ha

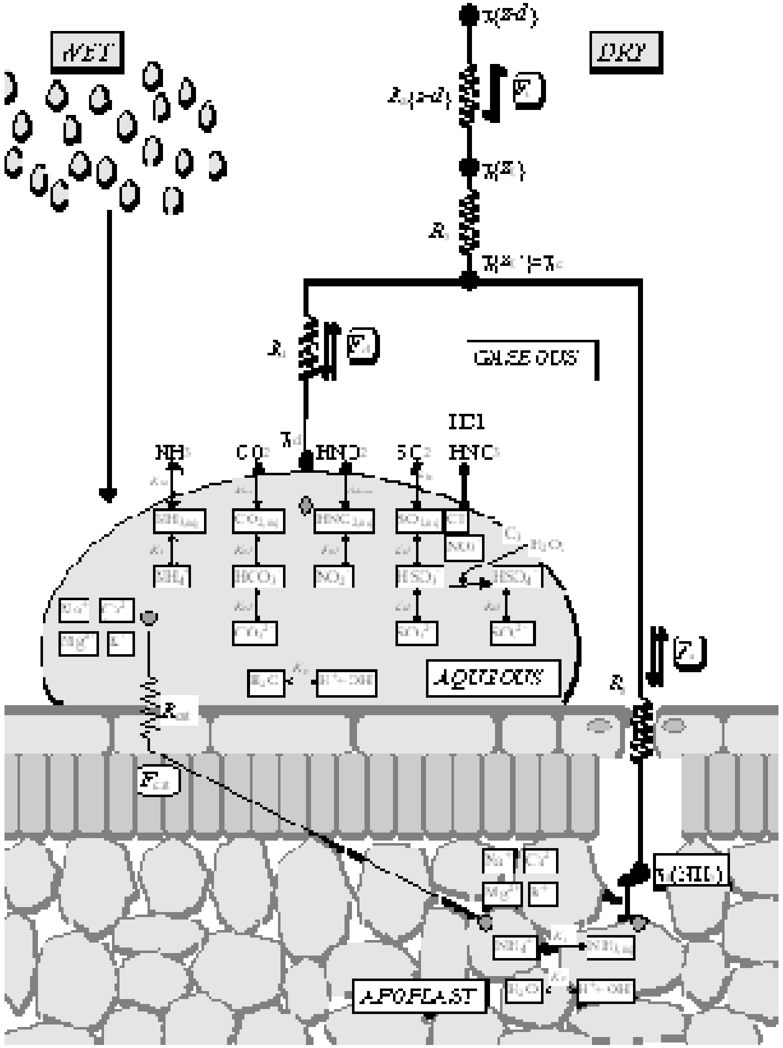
SO_2 deposition on vegetation



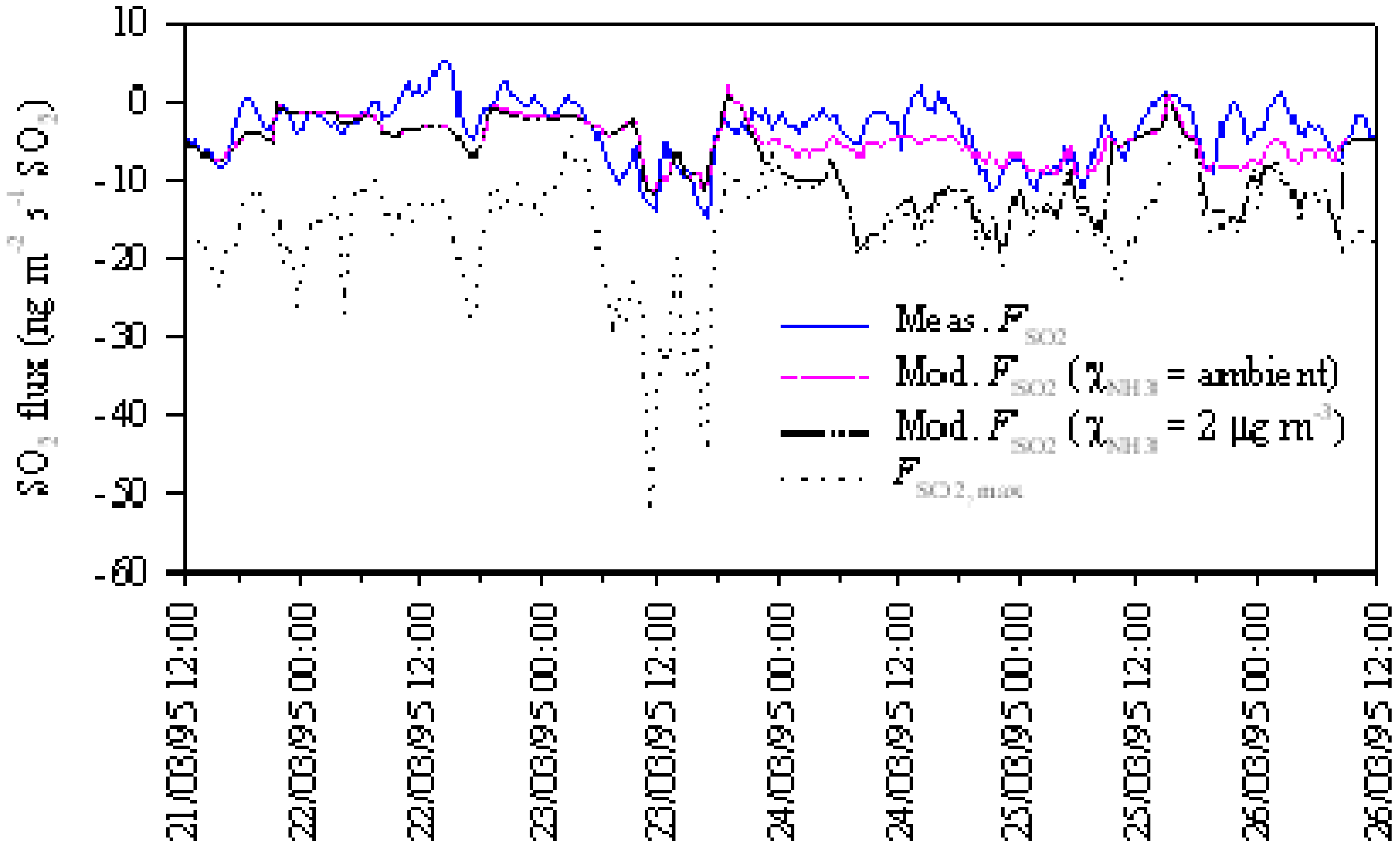
SO_2 deposition on vegetation



Leaf surface chemistry model for SO_2 deposition

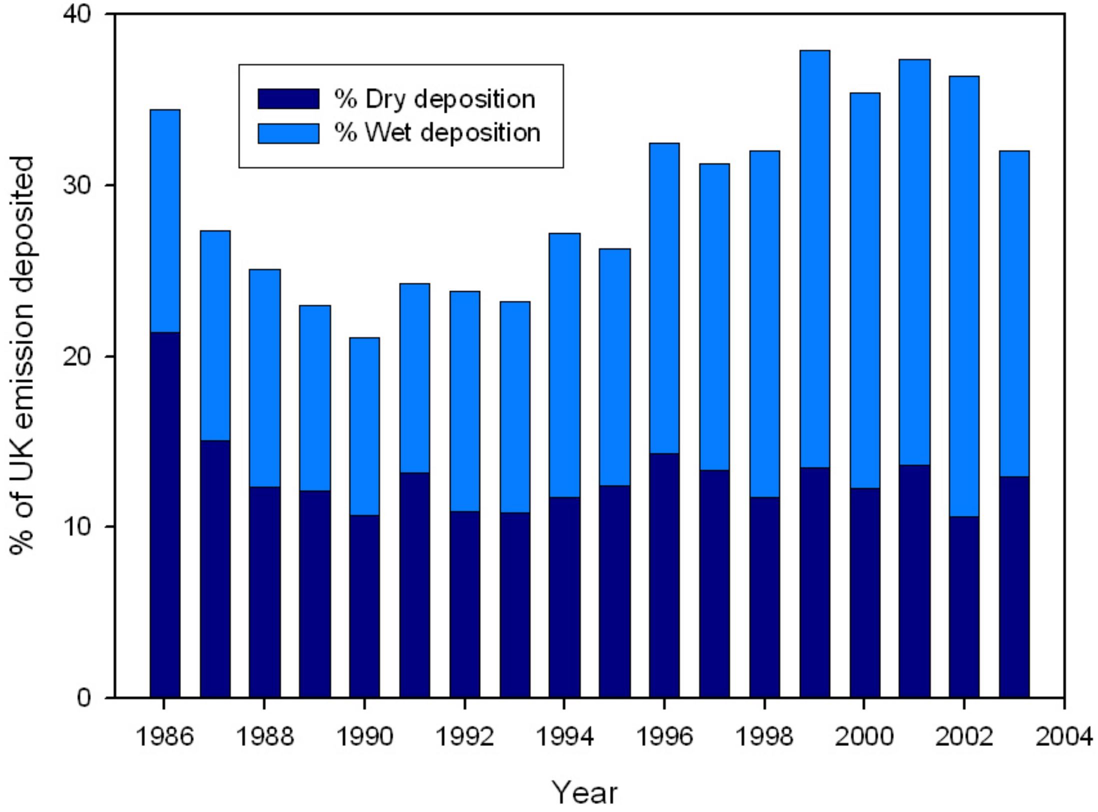


Model- measurement comparison



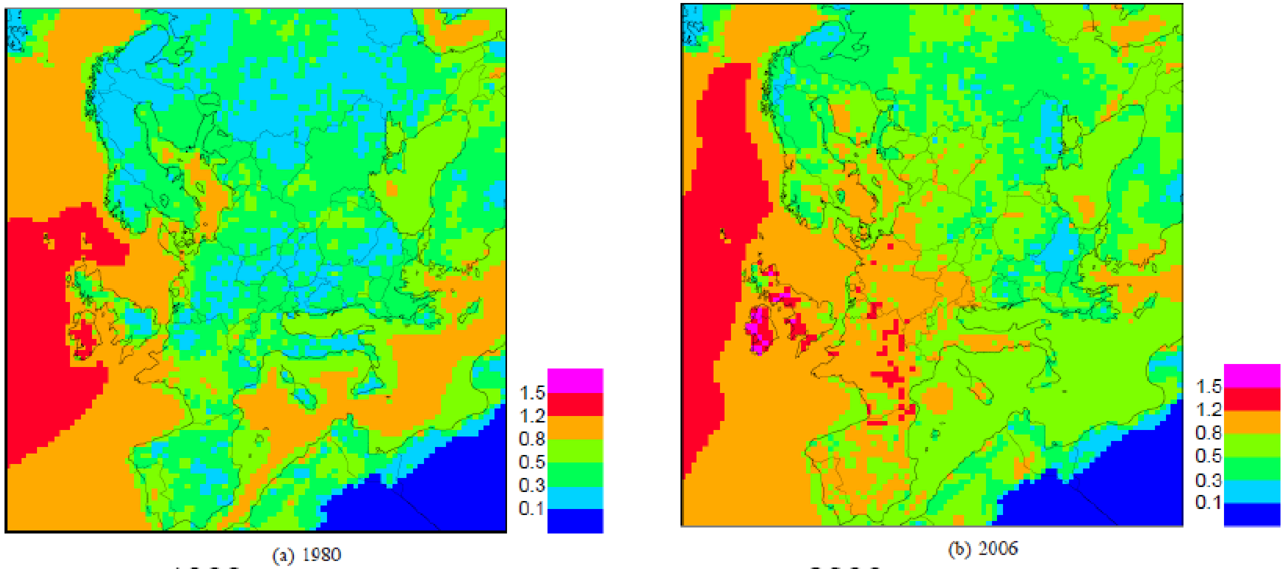
Long-term trends in dry deposition

% of UK emissions deposited in UK



Sulphur deposition and feedbacks

Co-deposition of SO_2 and NH_3



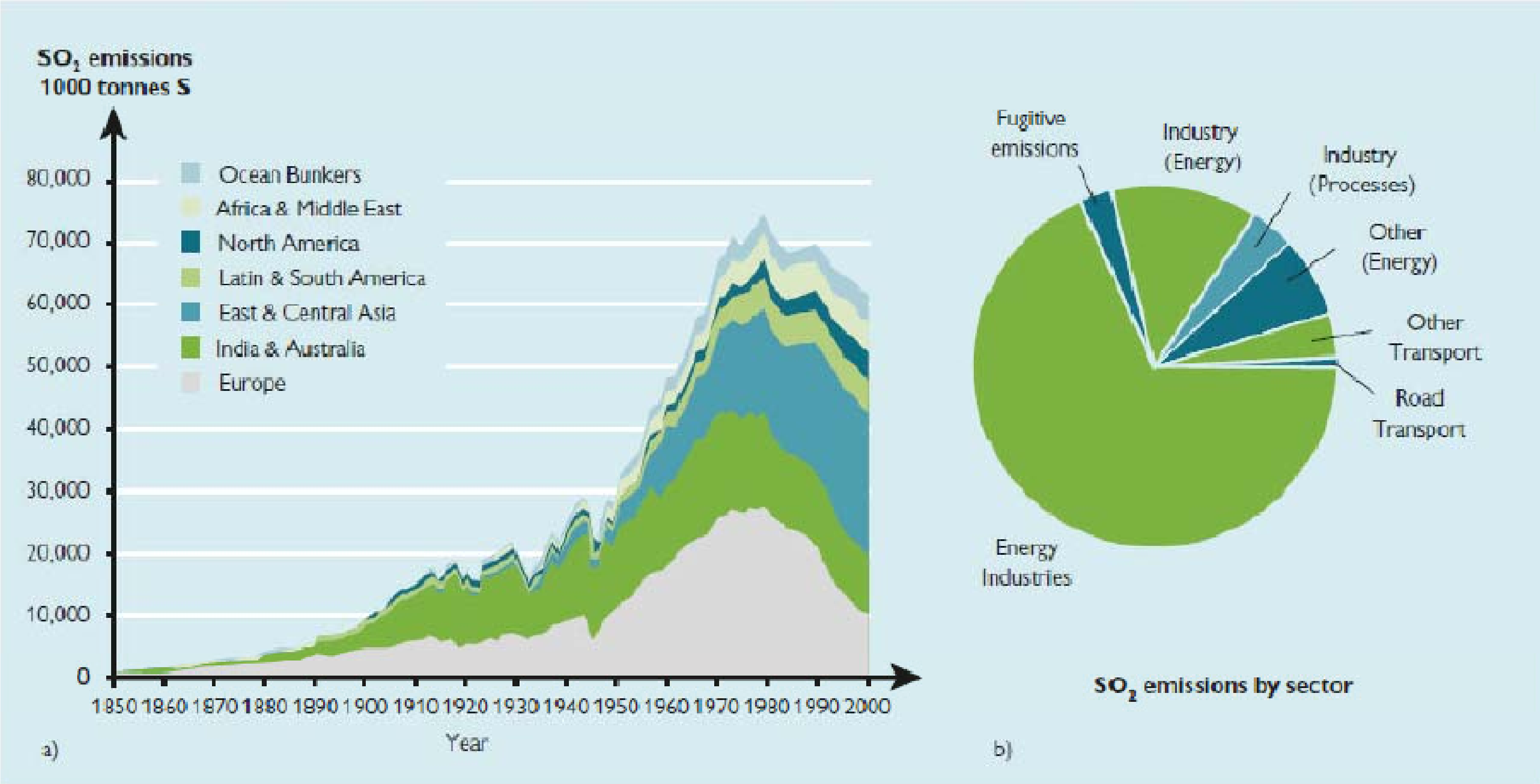
1980 2006

Modelled dry deposition velocities (cms^{-1}) of SO_2 over Europe for 1980 and 2006

Increasing dry deposition velocity of SO2 – less SO2 available for sulphate formation – less negative RF from aerosols – positive feedback

(Hilde Fagerli et al. in preparation, EMEP MSCW, 2009)

Global SO_2 emissions by region, 1850–2000



Source: a) Smith et al., 2004; b) EU-27 EEA

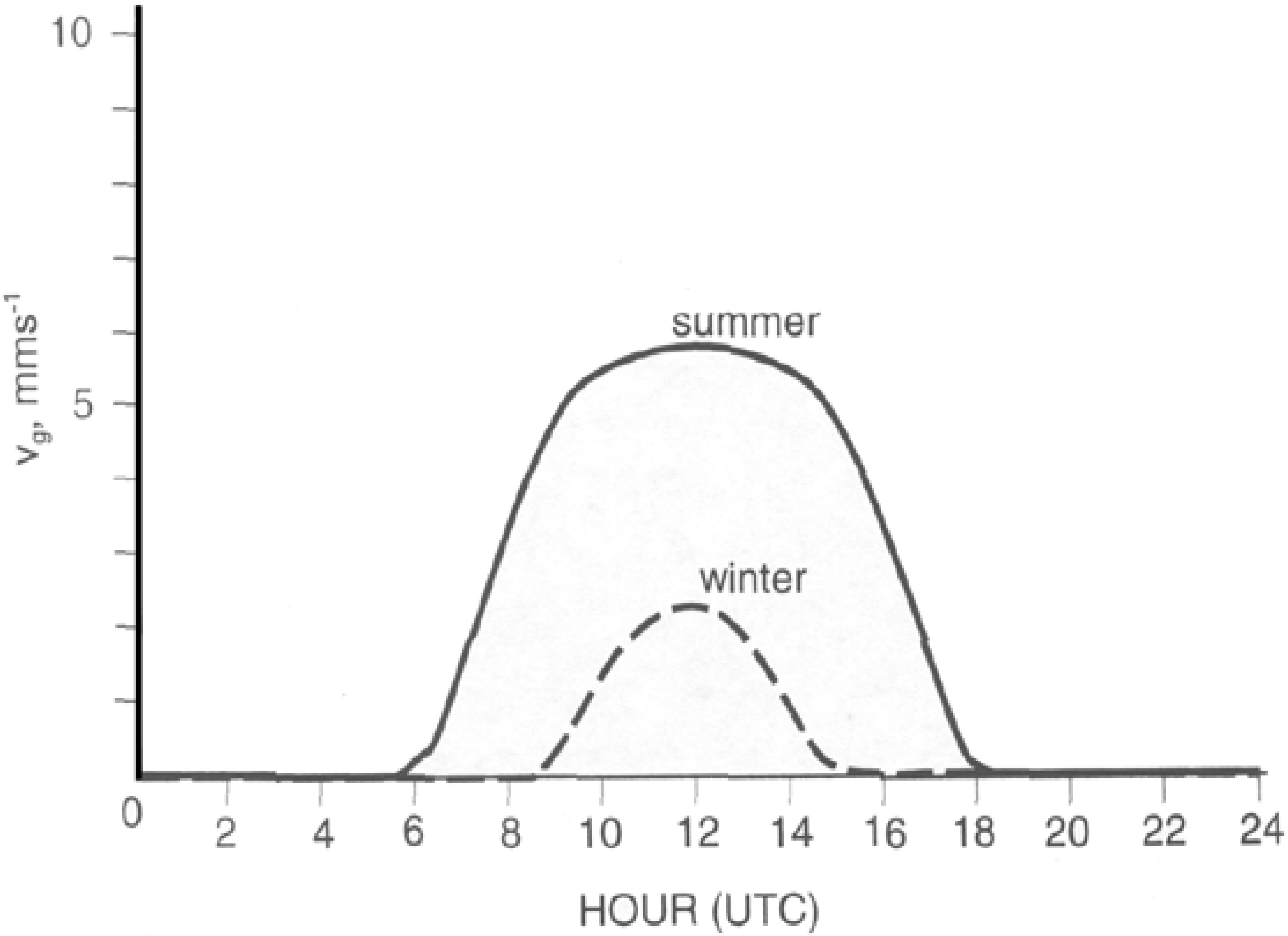
Reactive nitrogen compounds

- **Oxidized Nitrogen NO, NO2, HNO3, HONO, PAN**

- Processes in soils
- Compensation points
- Canopy- atmosphere interactions and a Discussion of the canopy uptake of the fraction of soil NO emission converted to NO2 and taken up by the canopy.
- Up-scaling of NO emission to regions, continents and global scales and their uncertainties.
- HNO3 deposition, the presence of a surface resistance
- Uptake of NO2 by vegetation and soil.
- Formation of HONO at terrestrial surfaces

NO_2 deposition on vegetation

Diurnal cycle in NO_2 deposition velocity.

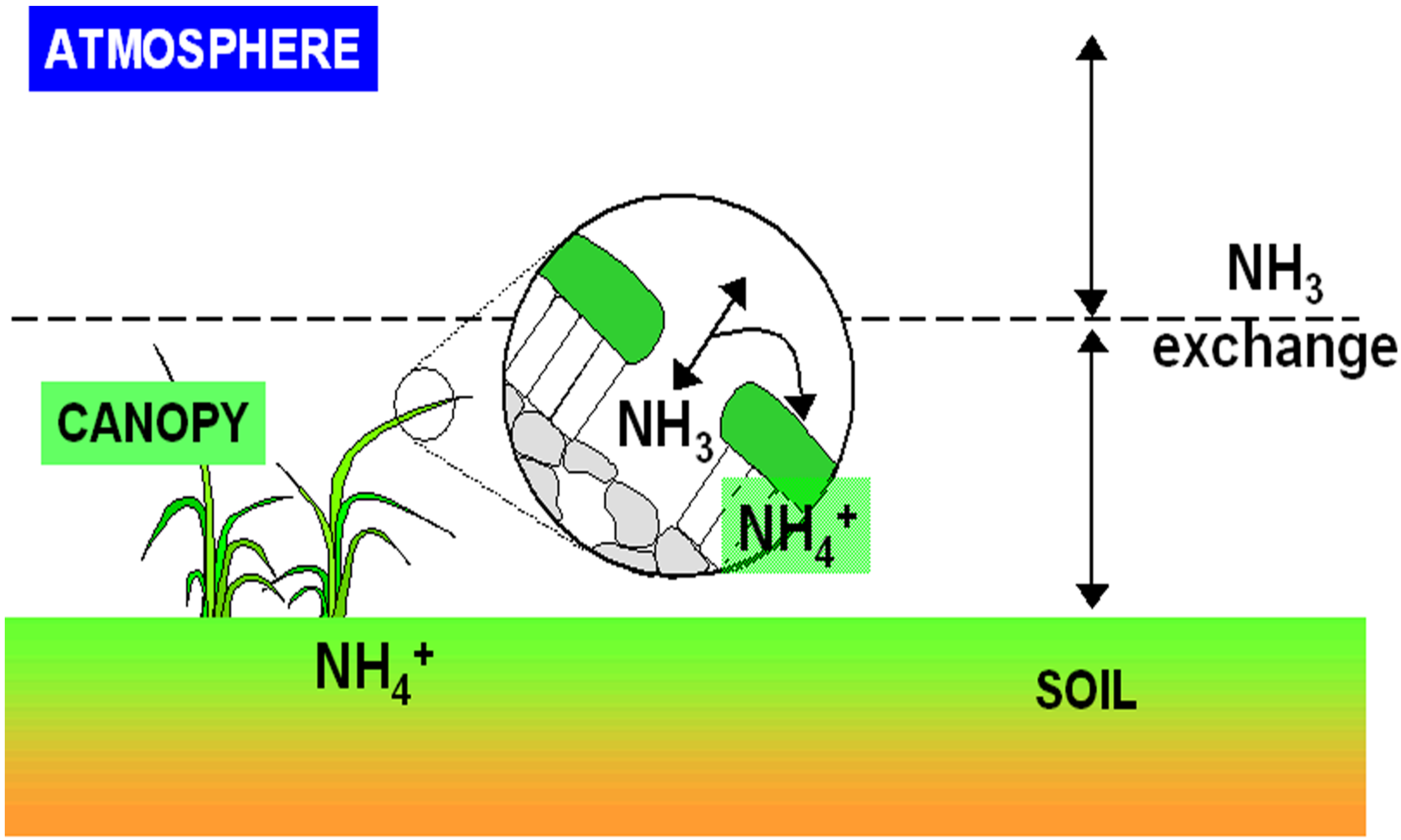


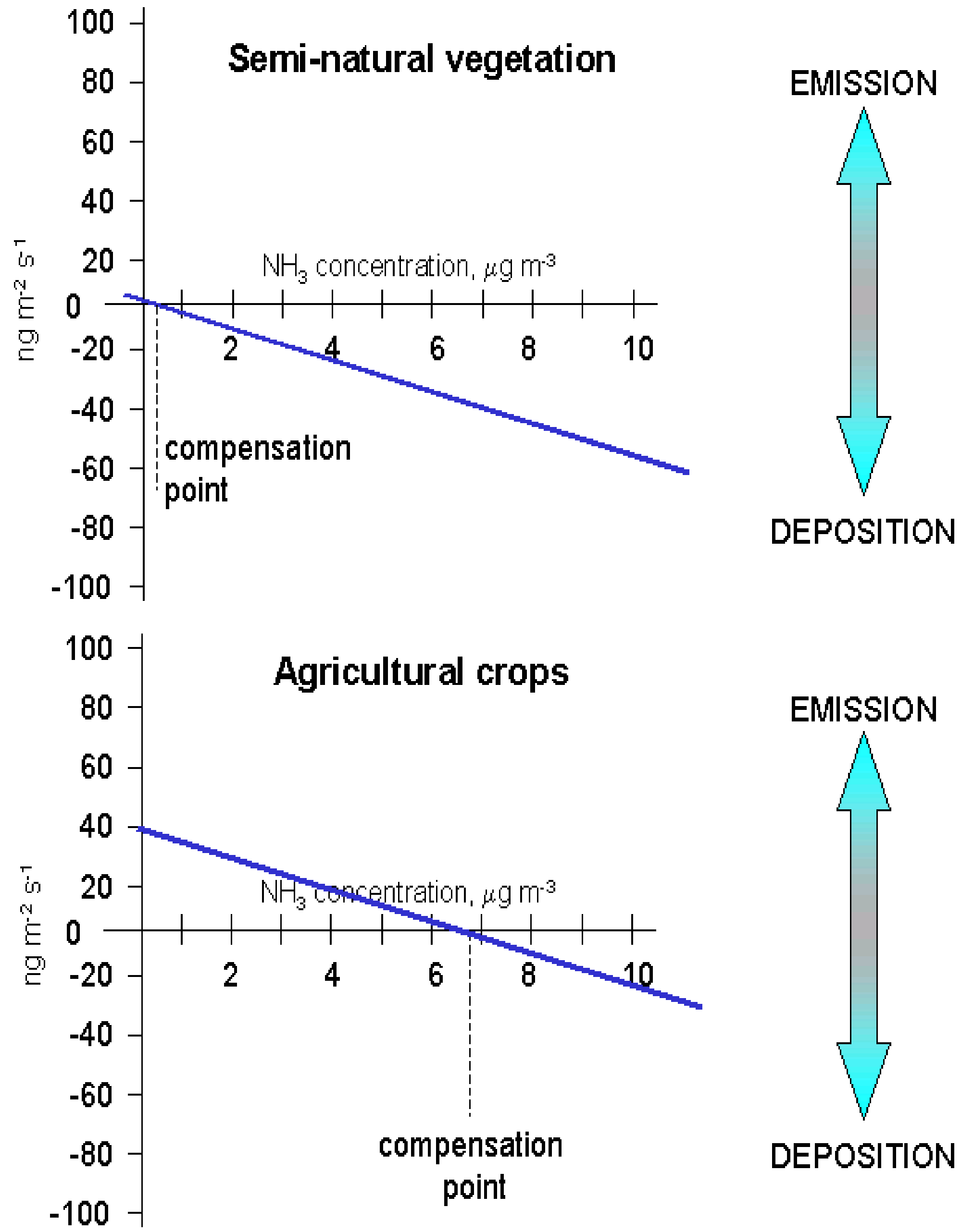
Ammonia

Reduced Nitrogen (NH_3)

- Processes at the surface regulating NH4+ concentrations in the apoplast, and their interaction with the atmosphere and external (leaf surface) factors.
- Compensation points
- Concentration dependent canopy resistances for NH_3 deposition
- Up-scaling to the canopy, landscape, region and continent.

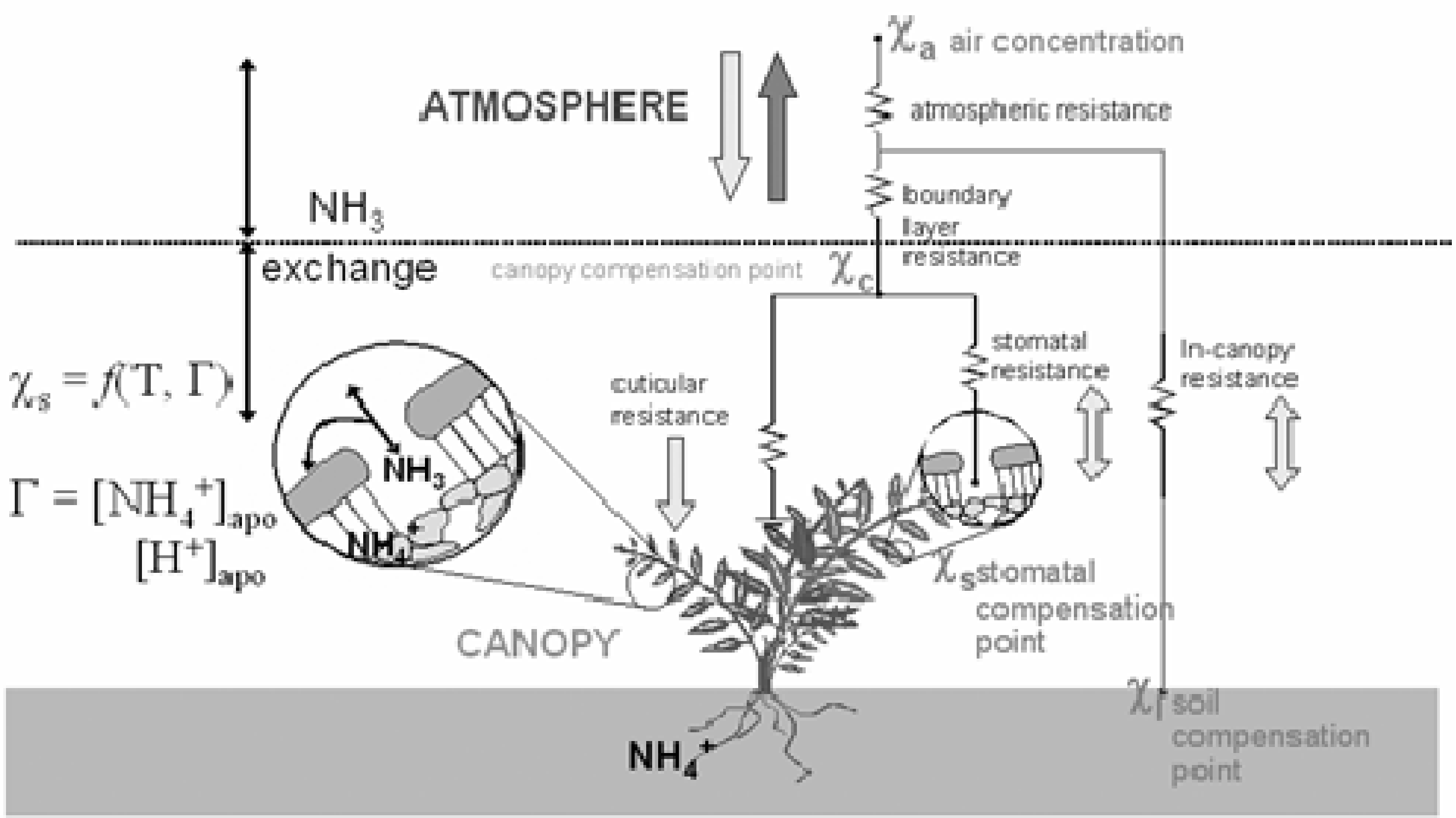
Bi-directional exchange





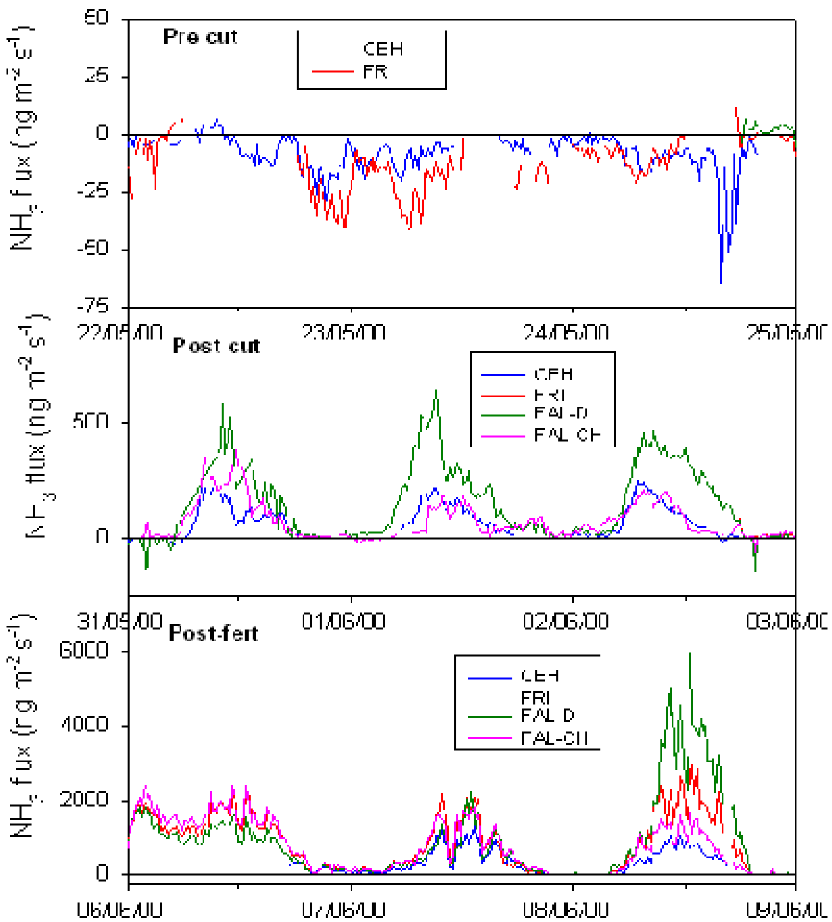
Semi-natural vegetation
EMISSION
DEPOSITION
ng m-2 s-1
NH3 concentration, µg m-3
100
80
60
40
20
0
-20
-40
-60
-80
-100
2
4
6
8
10
compensation point
Agricultural crops
EMISSION
DEPOSITION
ng m-2 s-1
NH3 concentration, µg m-3
100
80
60
40
20
0
-20
-40
-60
-80
-100
2
4
6
8
10
compensation point

Canopy compensation point model

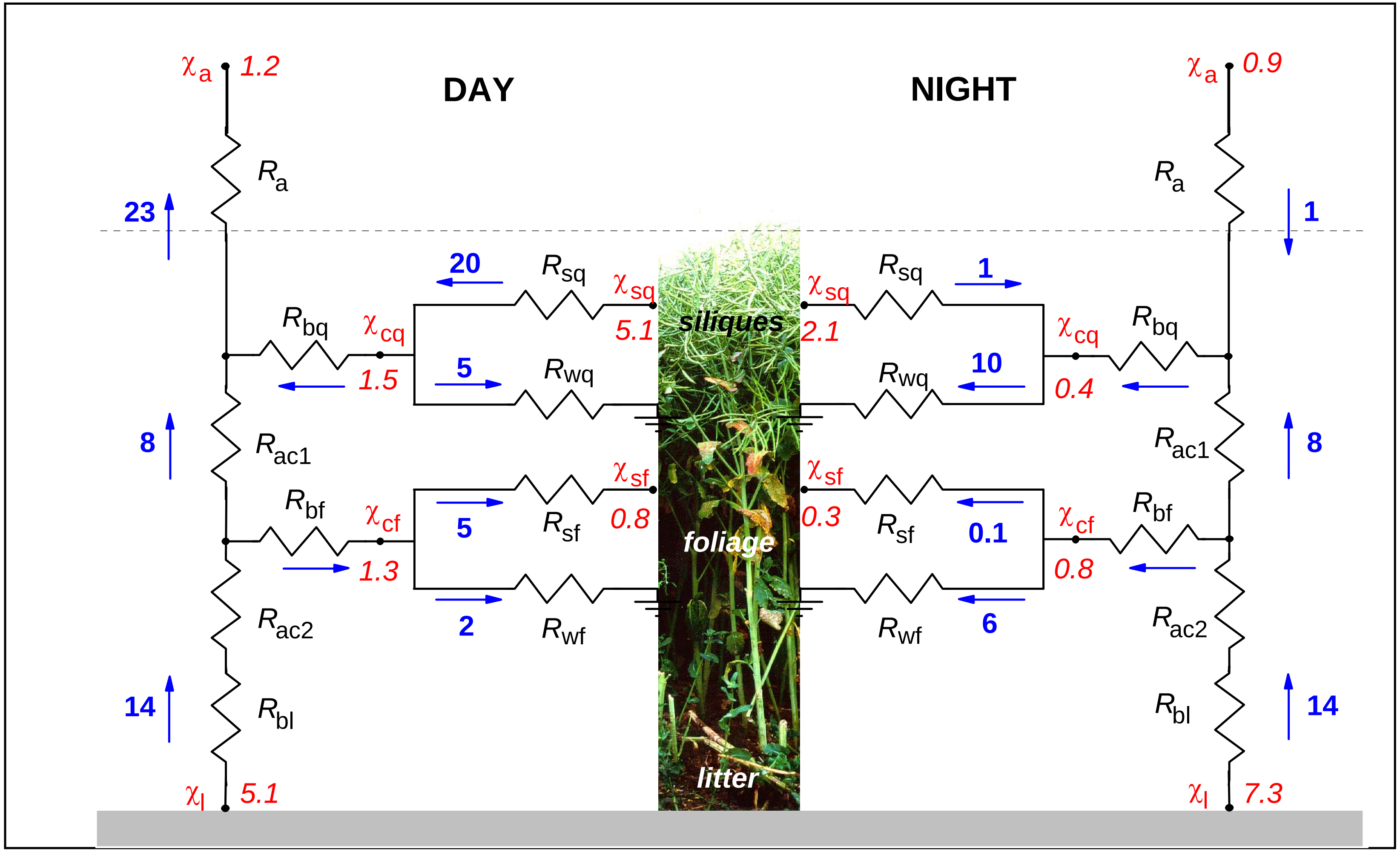


Ammonia exchange over vegetation

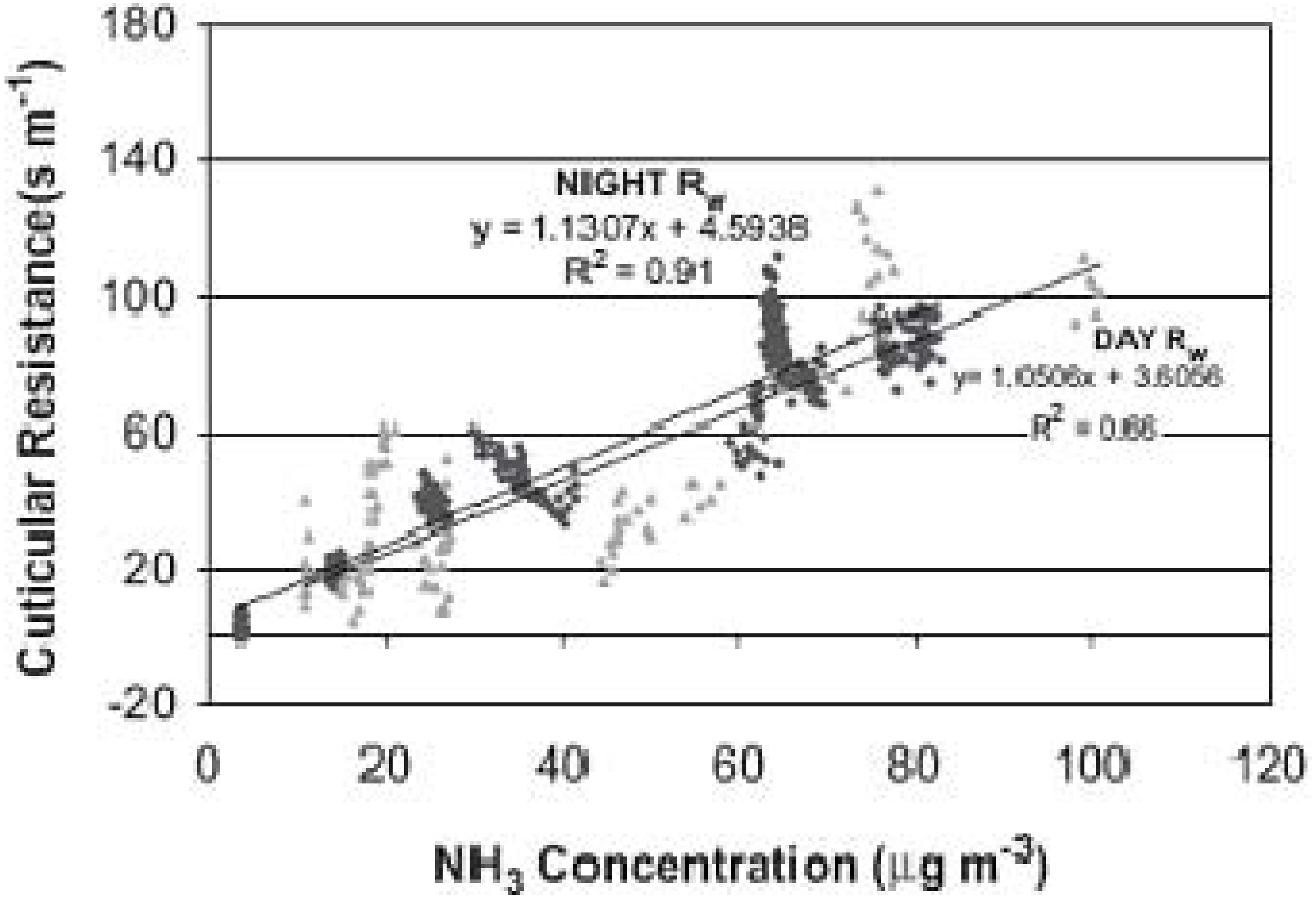
Intercomparison of methods to measure canopy-atmosphere exchange fluxes of NH_3 at Braunschweig over agricultural grassland



Resistance modelling of component fluxes of ammonia within plant canopies



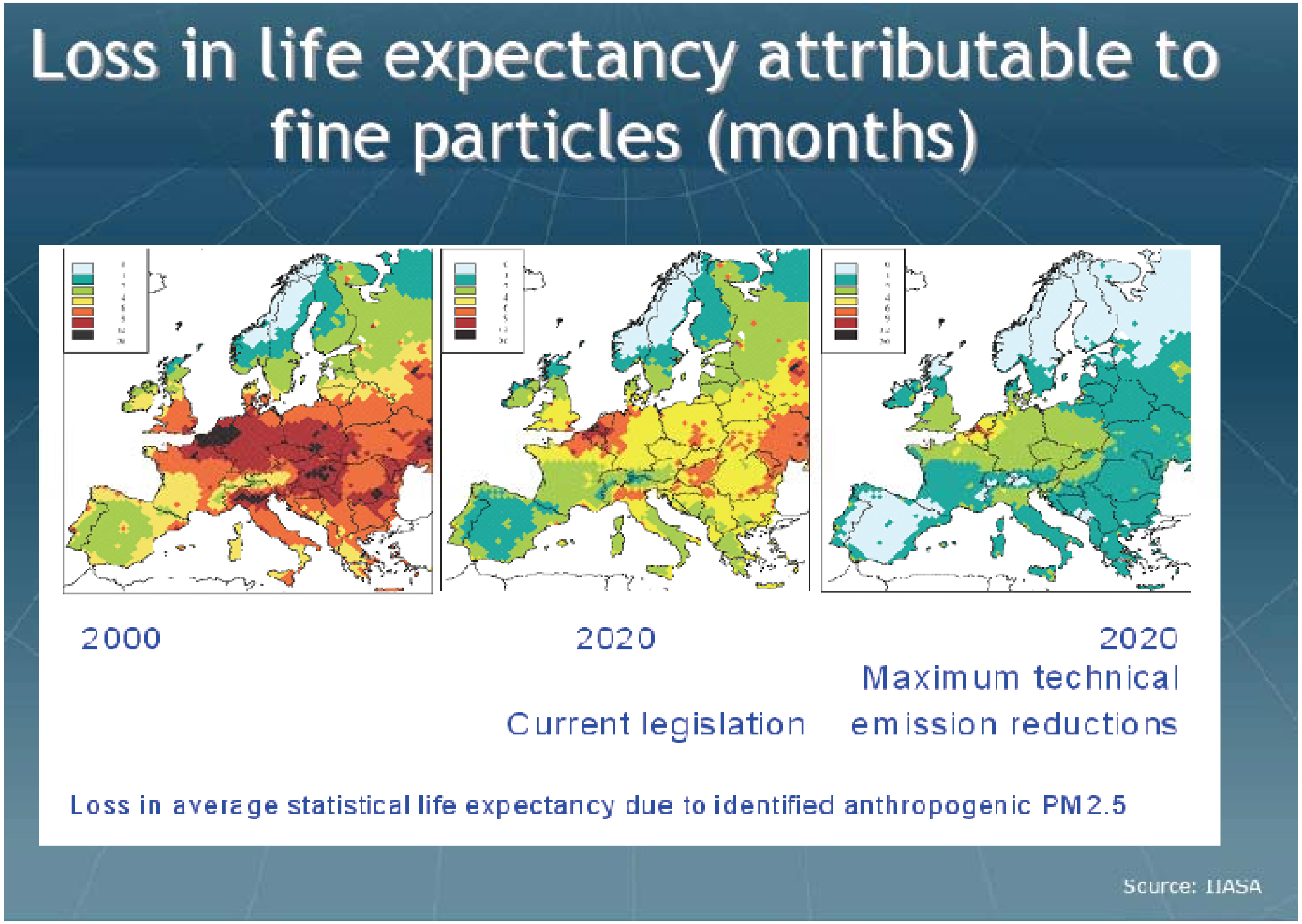
Concentration dependence of leaf surface uptake of ammonia



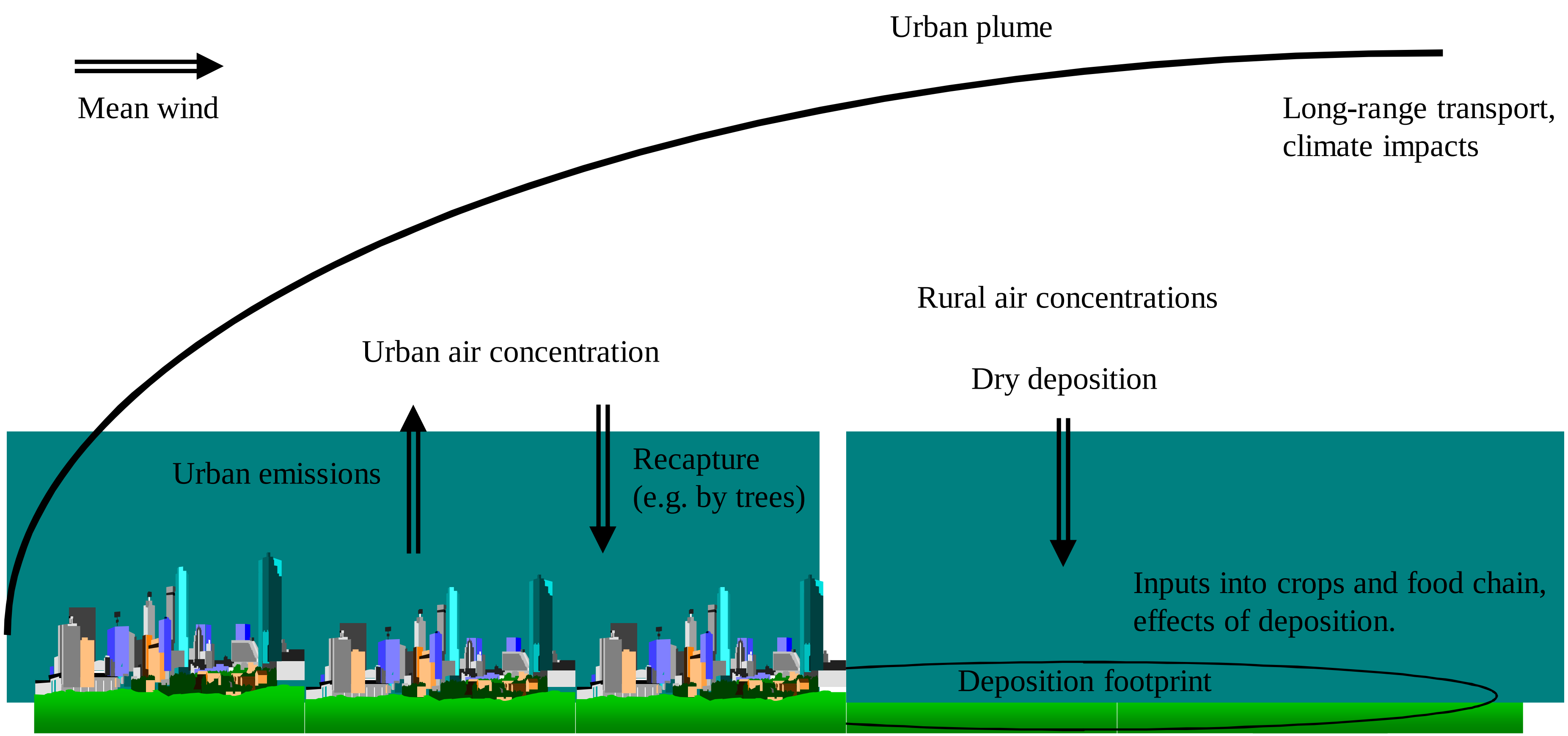
Aerosols

Particulate matter

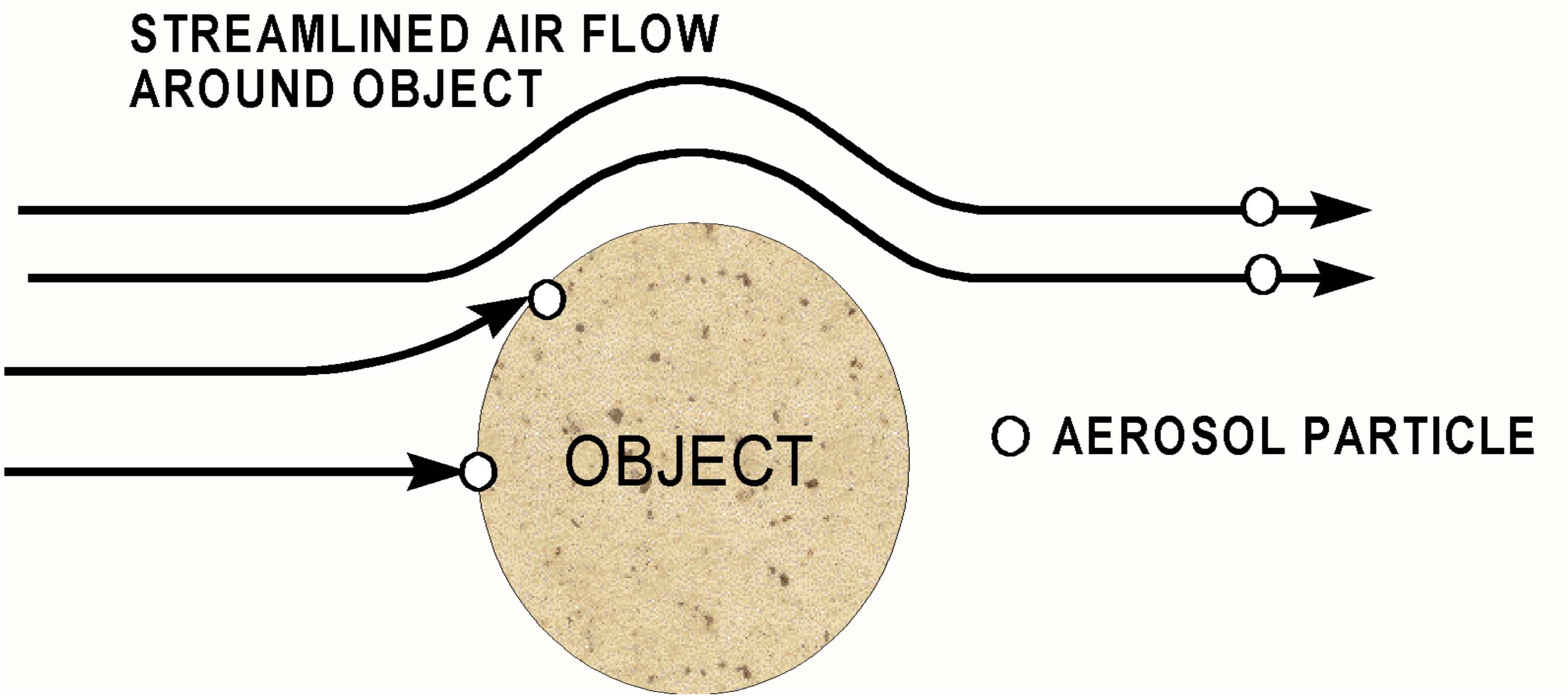
Particle emission and deposition



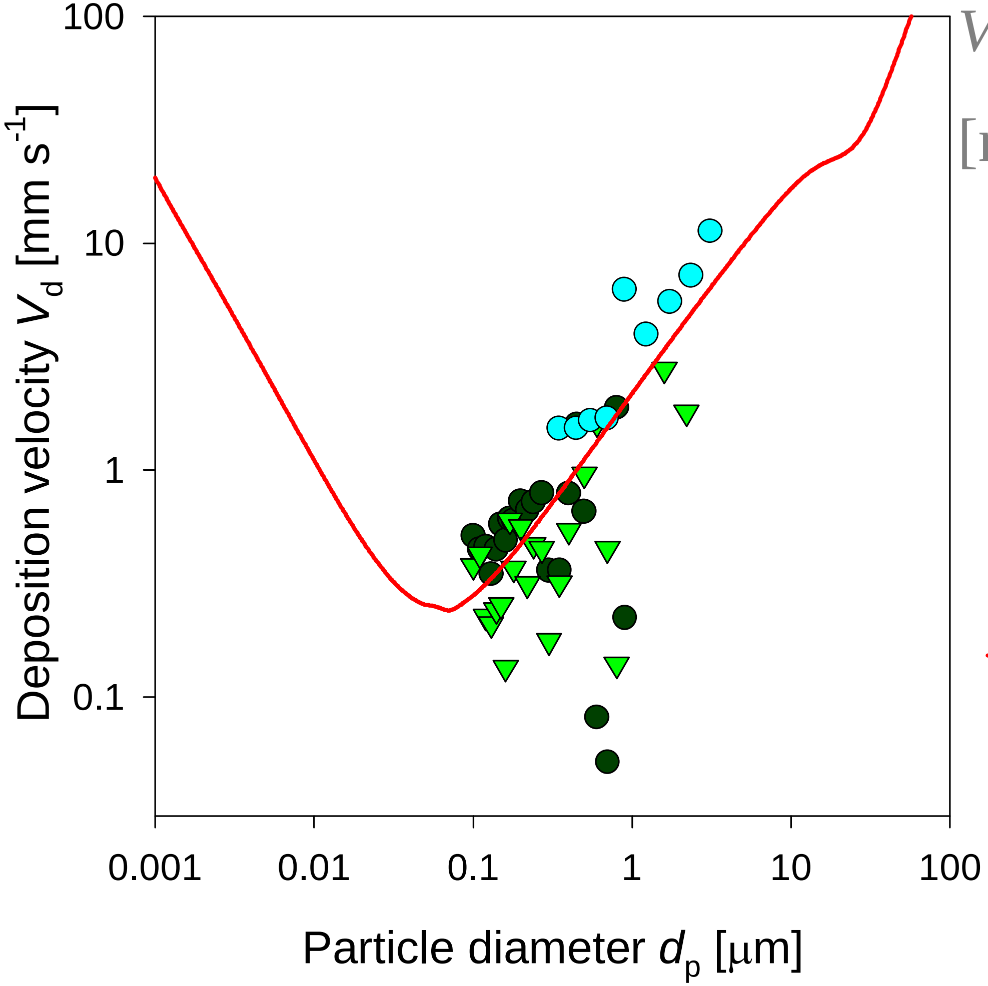
Emission, recapture and deposition



AEROSOL IMPACTION

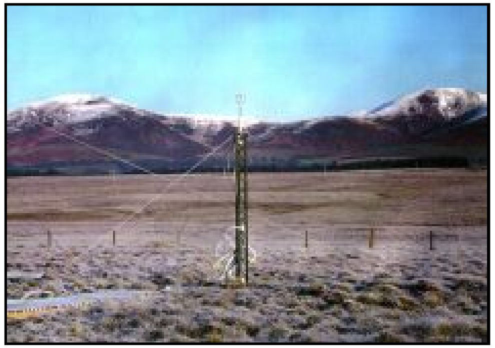


Aerosol deposition velocities as a function of size to moorland vegetation

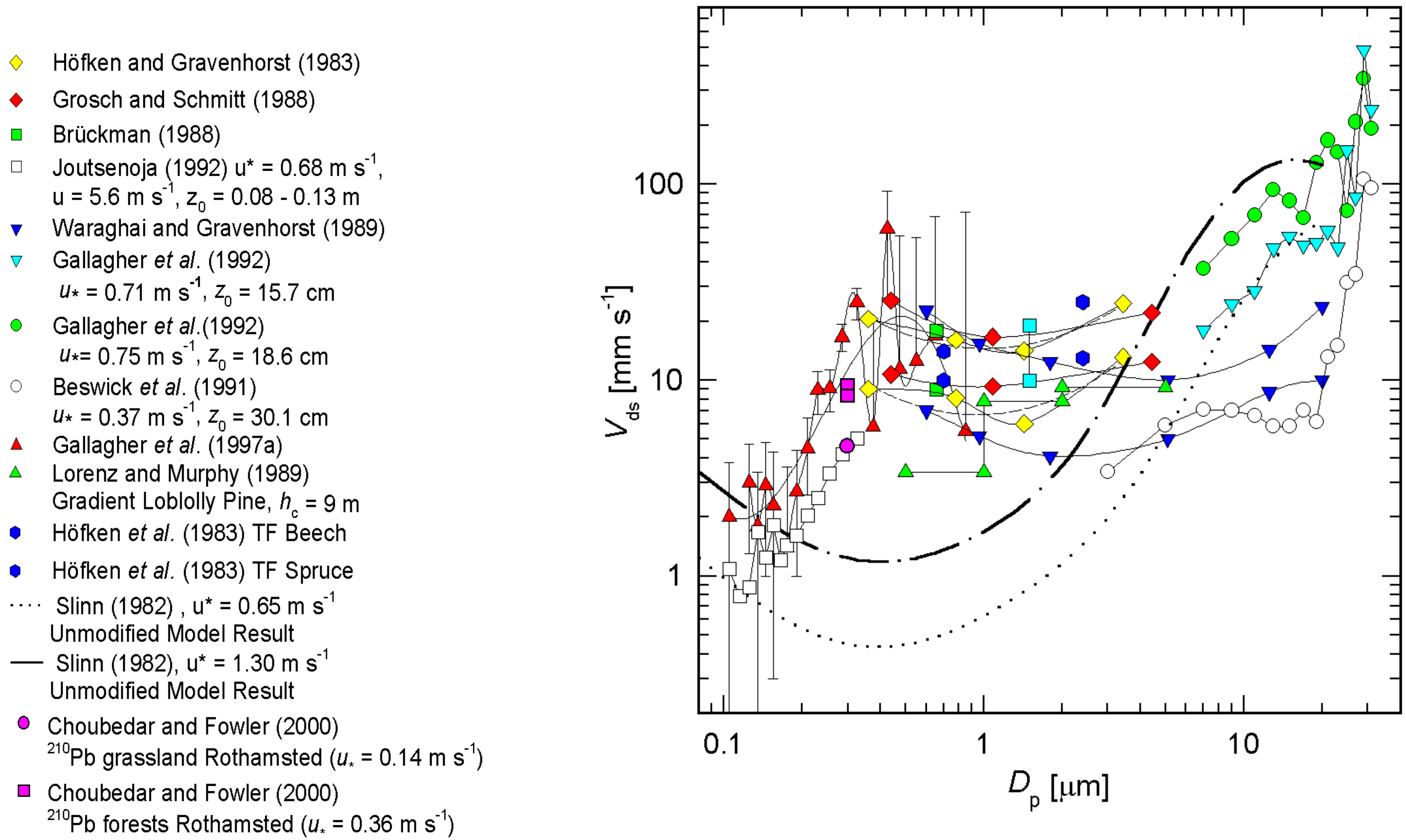


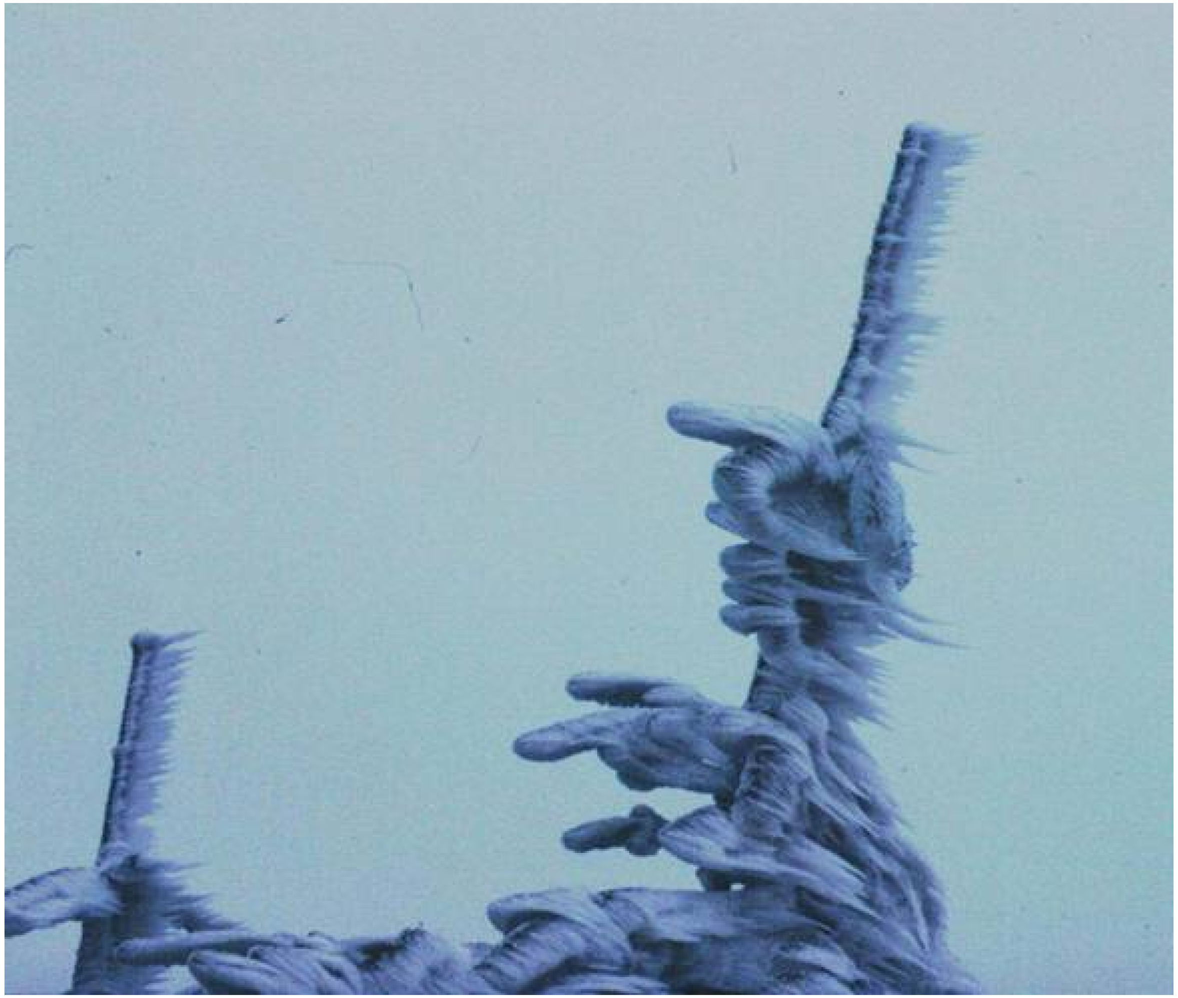
V_d = - flux / concentration

[m s^{-1}] = [# m^{-2} s^{-1}] / [# m^{-3}]



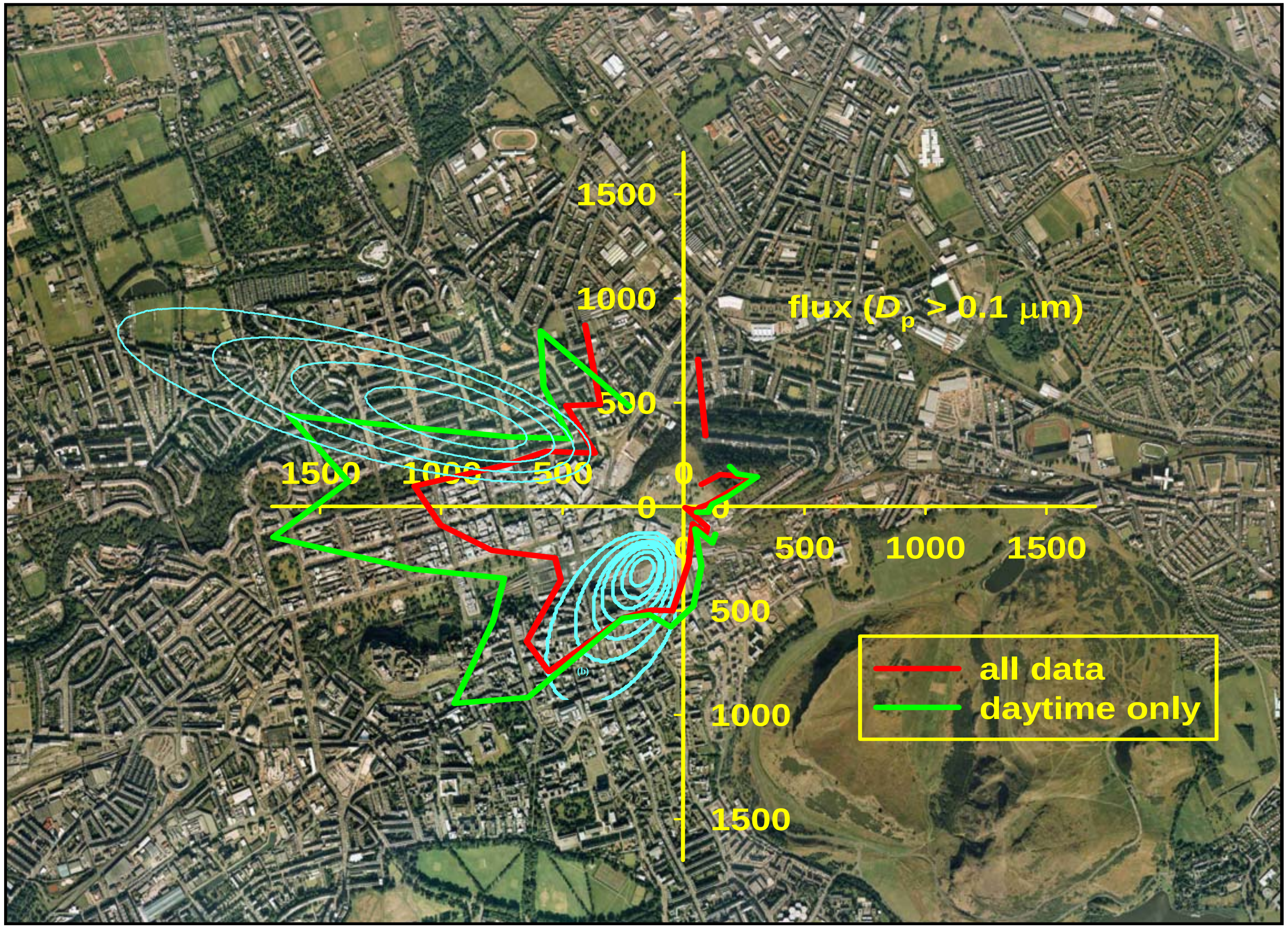
Aerosol deposition velocities as a function of size to trees





Measurements at the Nelson Monument

Example footprints and wind sector dependence of the flux

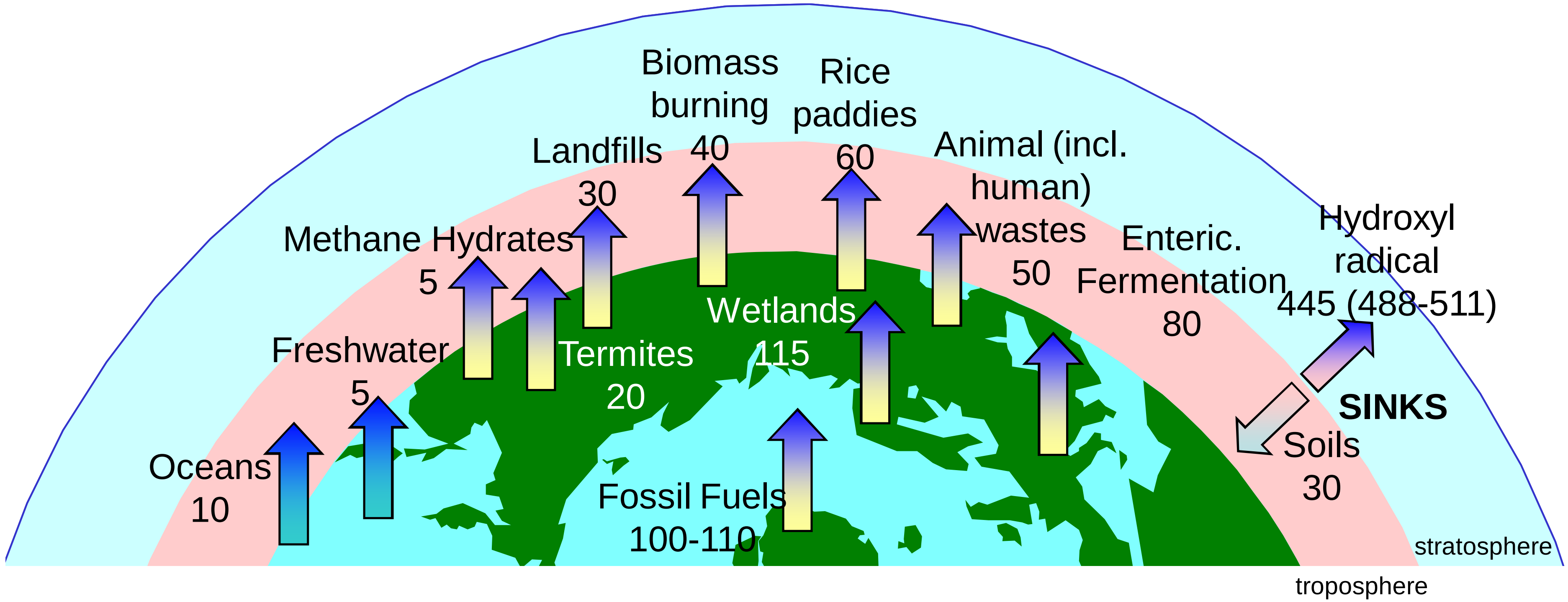


Probably stop here!

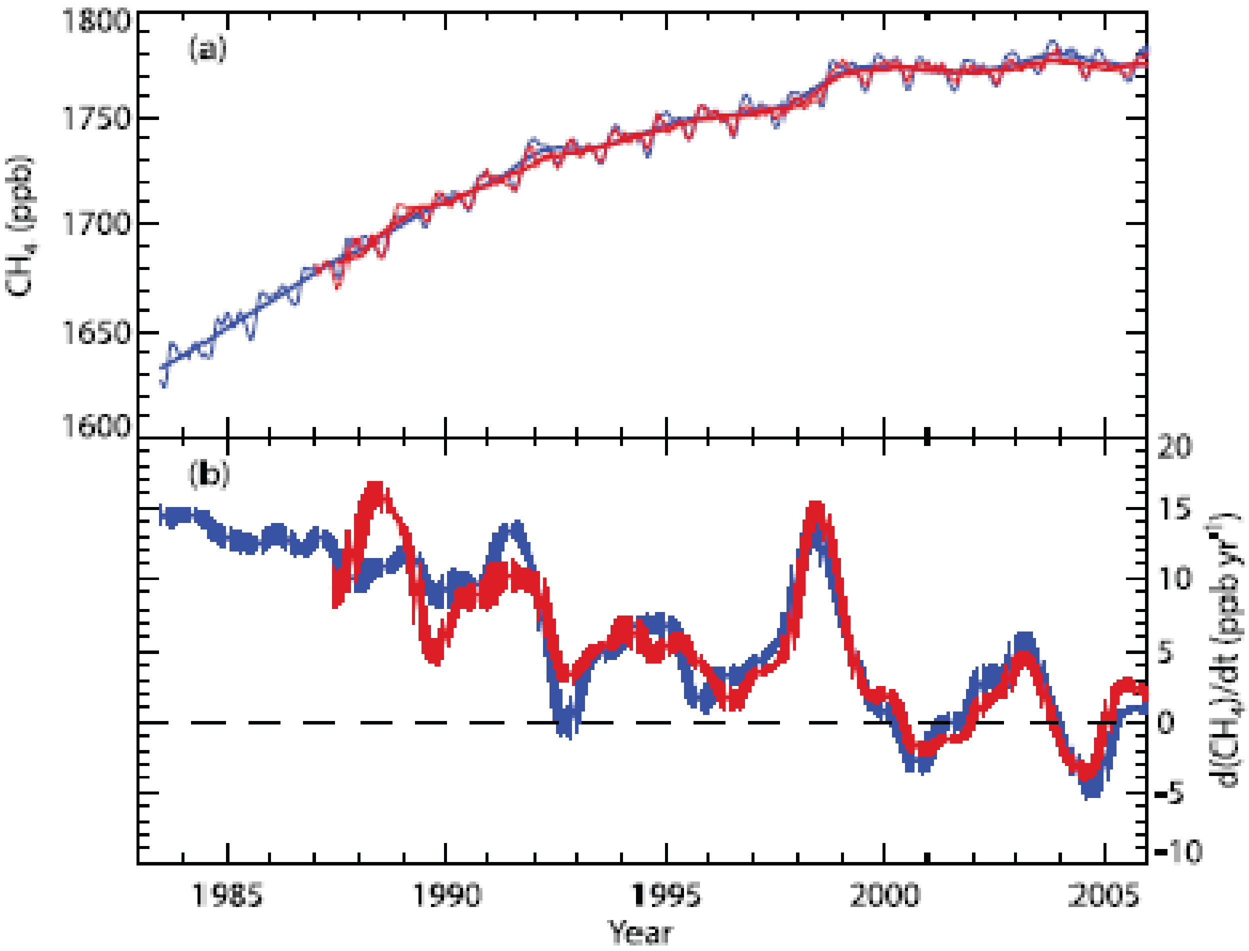
Methane

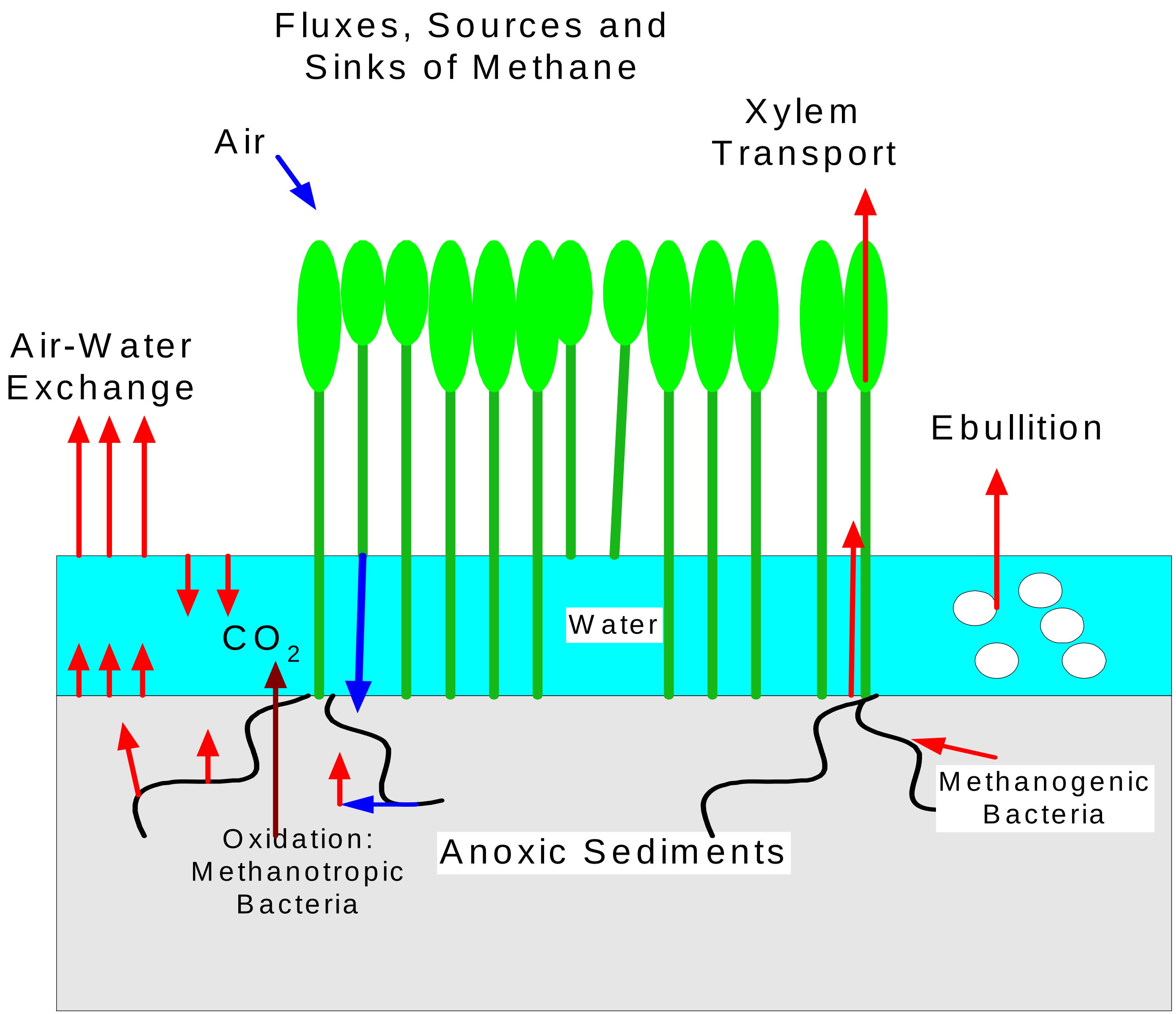
- Bi-directional,
- methane uptake by bacteria in aerobic soils
- Methane emission (by methanogenic bacteria) in anaerobic conditions.

GLOBAL METHANE BUDGET (Mt)	
Annual Source	515 Mt
Annual Sink	475 Mt
Net Input to Troposphere	30 - 50 Mt

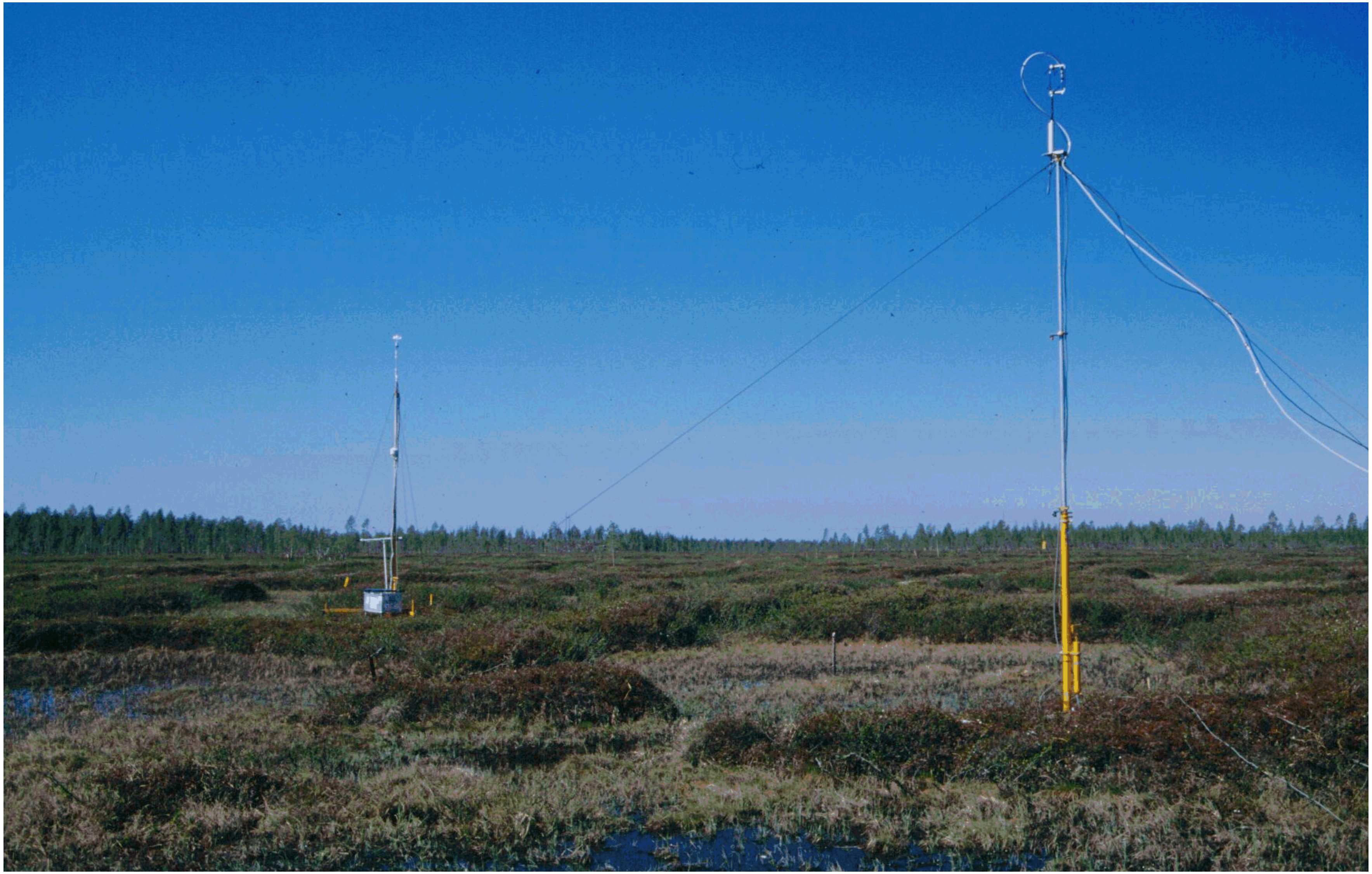


Greenhouse gas exchange and ecosystems: methane

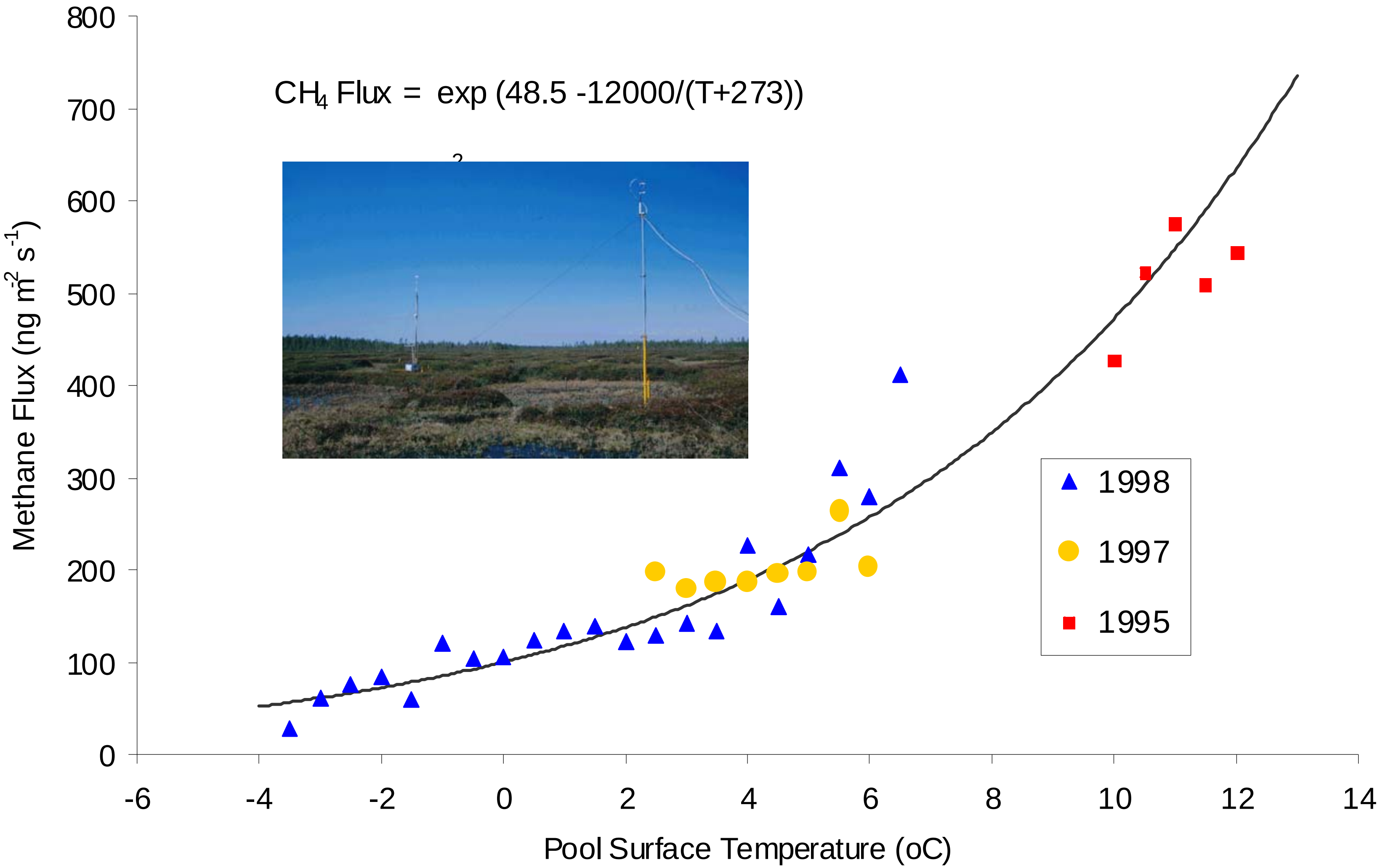



Fluxes, Sources and
Sinks of Methane
Air
Xylem
Transport
Air-Water
Exchange
Ebullition
CO_2
Water
Oxidation:
Methanotropic
Bacteria
Anoxic Sediments
Methanogenic
Bacteria

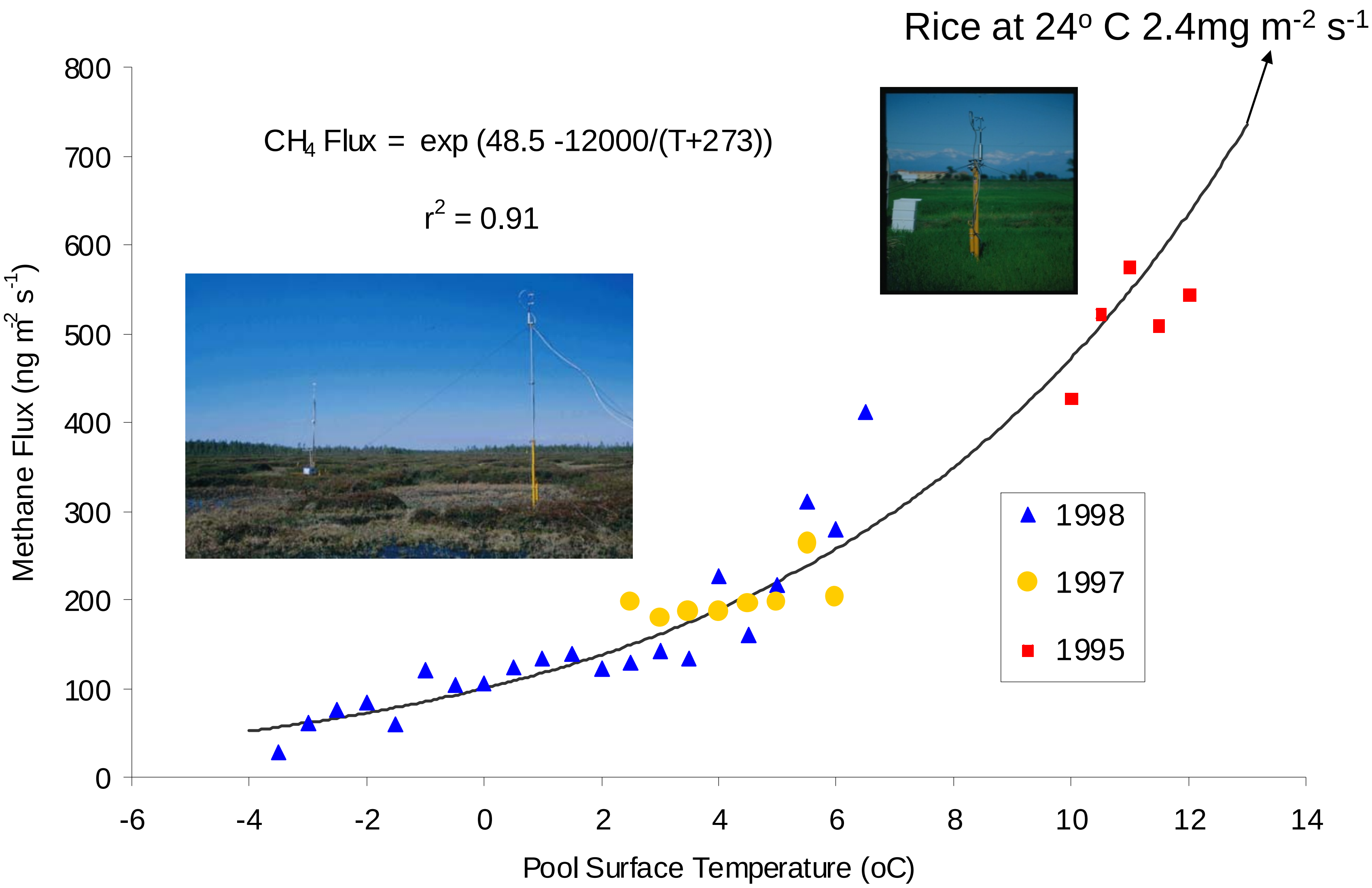
Lapland methane flux measurements



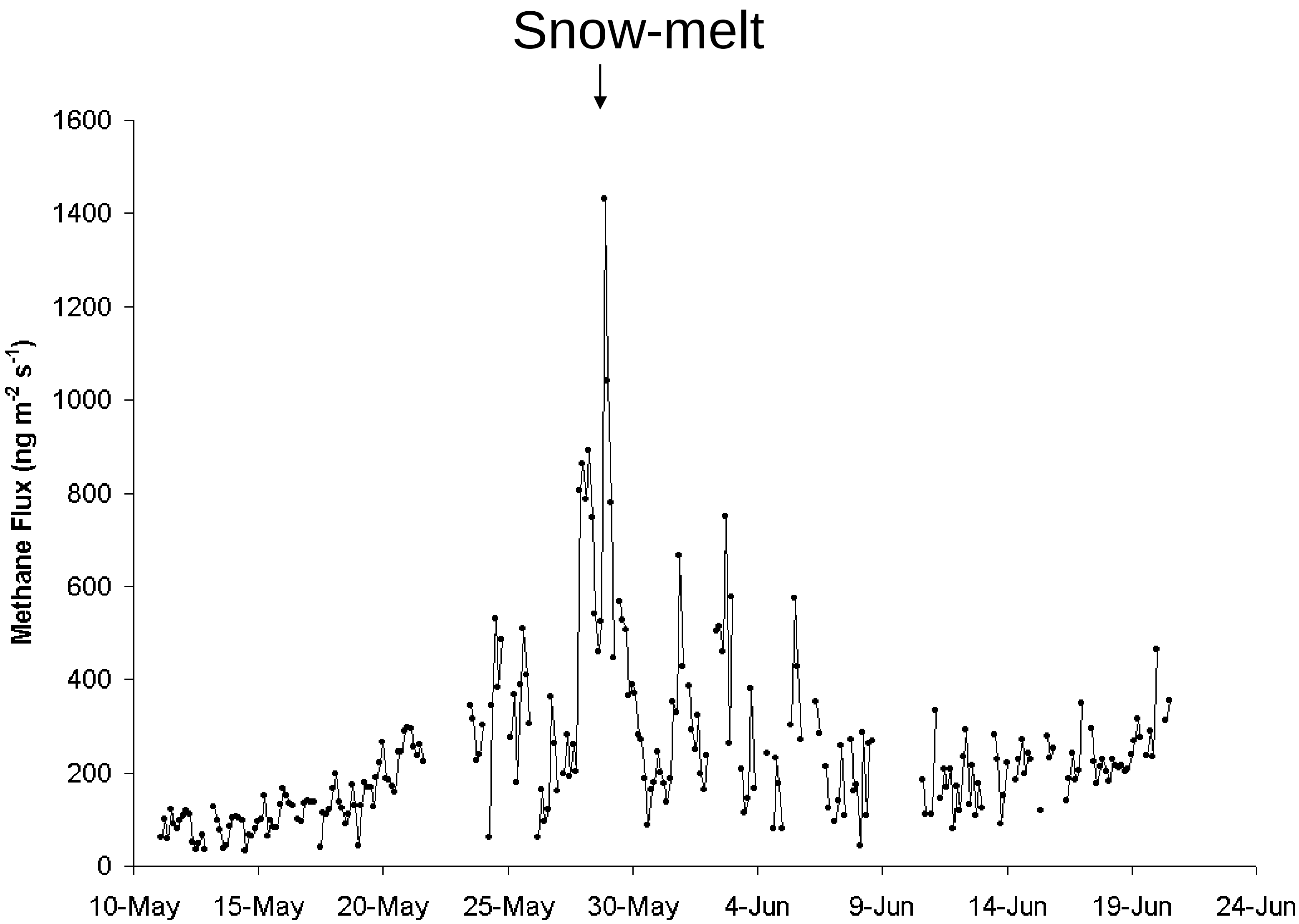

Source: CEH



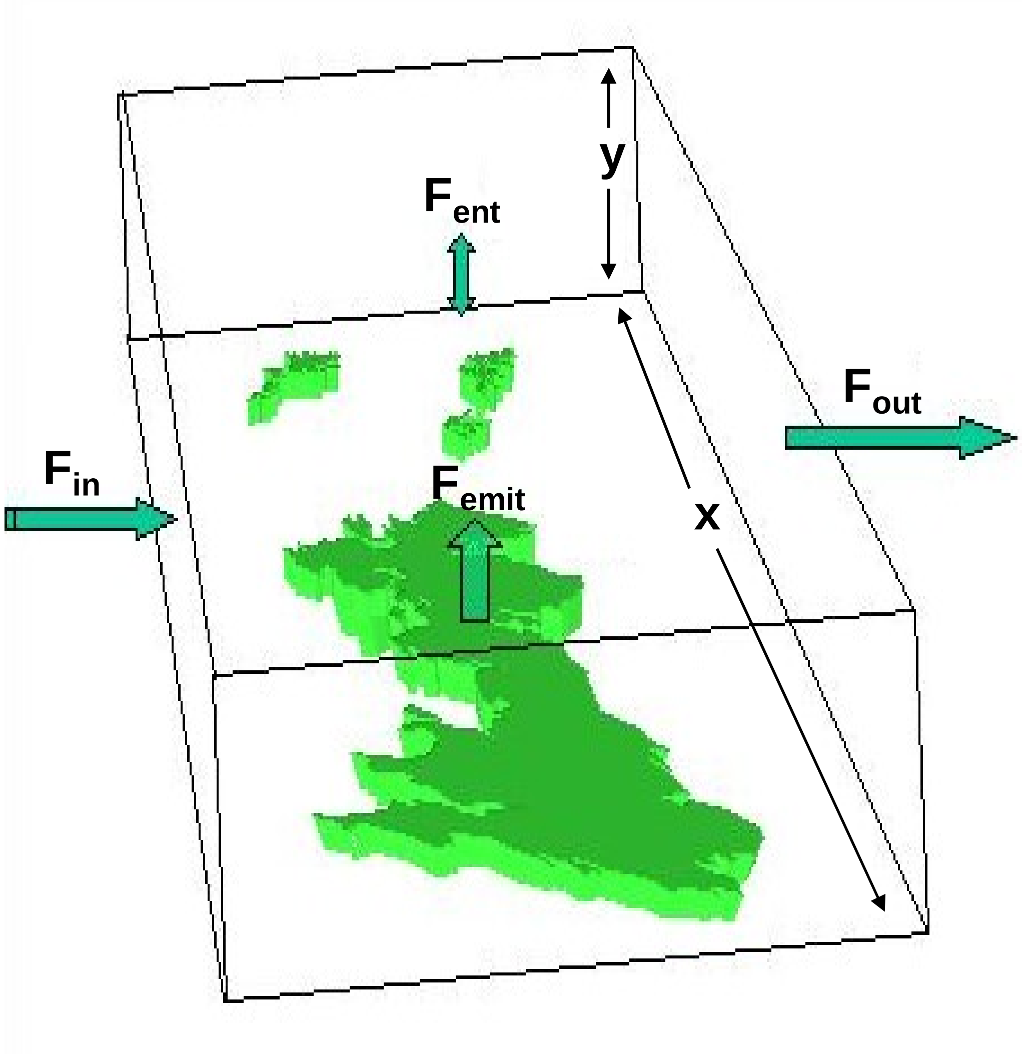
CH4 Flux = exp (48.5 -12000/(T+273))
Methane Flux (ng m-2 s-1)
800
700
600
500
400
300
200
100
0
-6
-4
-2
0
2
4
6
8
10
12
14
Pool Surface Temperature (oC)
1998
1997
1995


Rice at 24° C 2.4mg m^{-2} s^{-1}
CH_4 Flux = exp (48.5 -12000/(T+273))
r^2 = 0.91
Methane Flux (ng m^{-2} s^{-1})
800
700
600
500
400
300
200
100
0
-6
-4
-2
0
2
4
6
8
10
12
14
Pool Surface Temperature (oC)
1998
1997
1995

Methane emission during the snow-melt period in Lapland



Estimating Annual Emissions – Box Technique



- $F_{emit} = F_{out} - F_{in} - F_{ent}$
- $F_{in} = \iint u_{in}(y,z)c(y,z)dydz$
- $F_{out} = \iint u_{out}(y,z)c(y,z)dydz$
- $F_{ent = 0}$

F_{emit} = Flux emitted

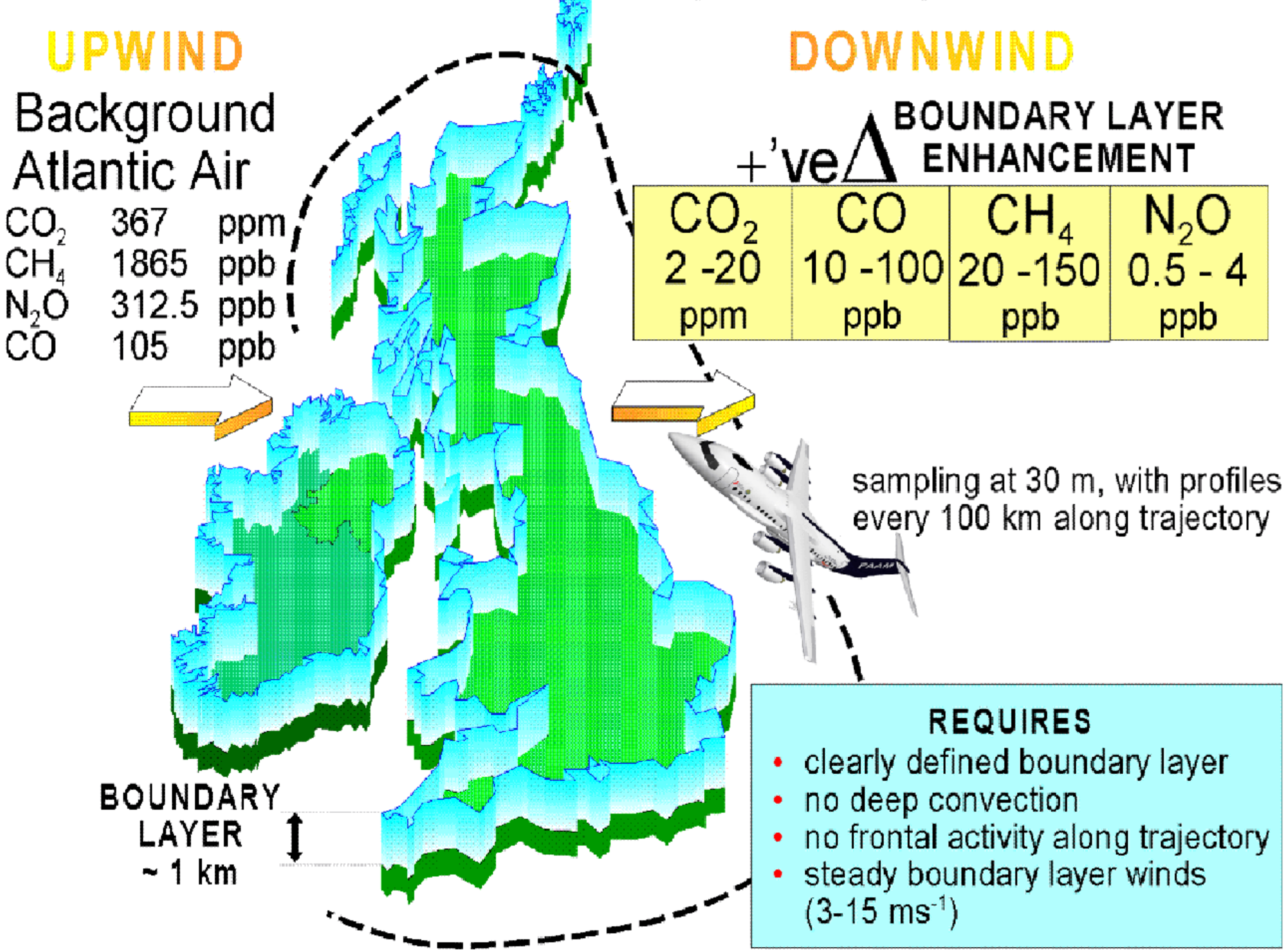
F_{in} = Flux in

F_{out} = Flux out

u = wind speed

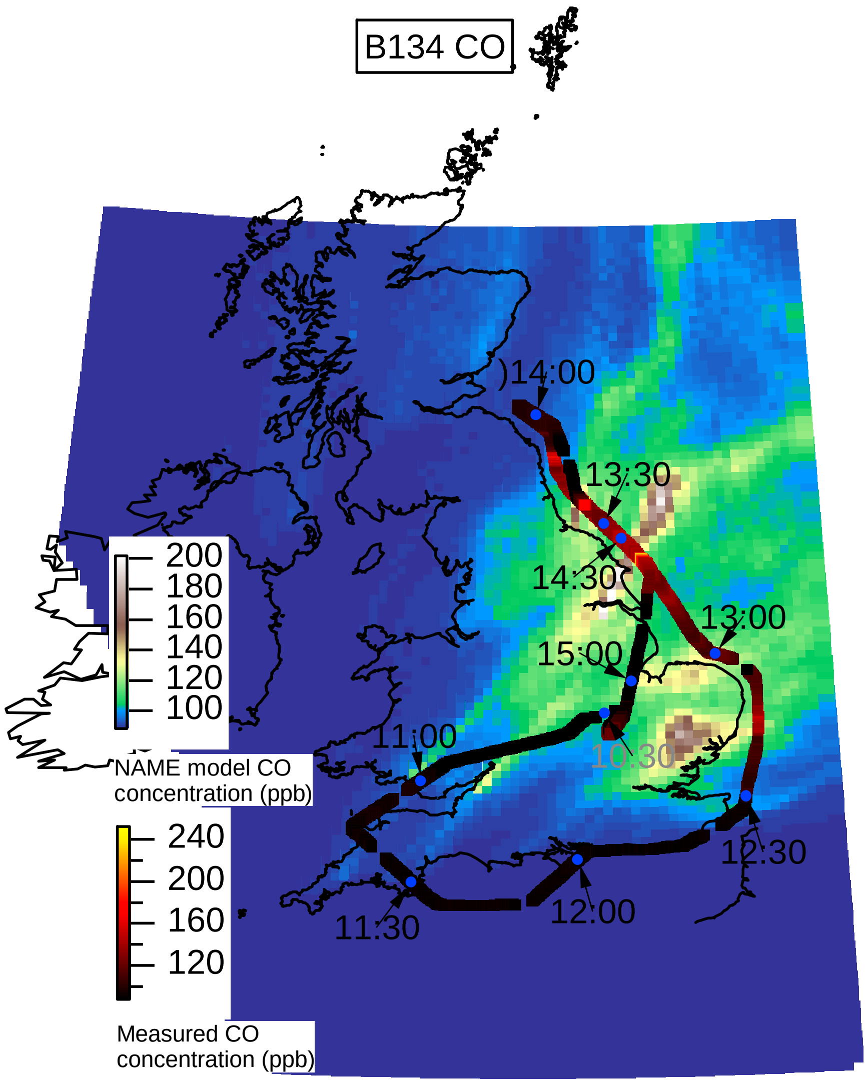
c = concentration

Atmospheric Mass Budgets at Regional Scales

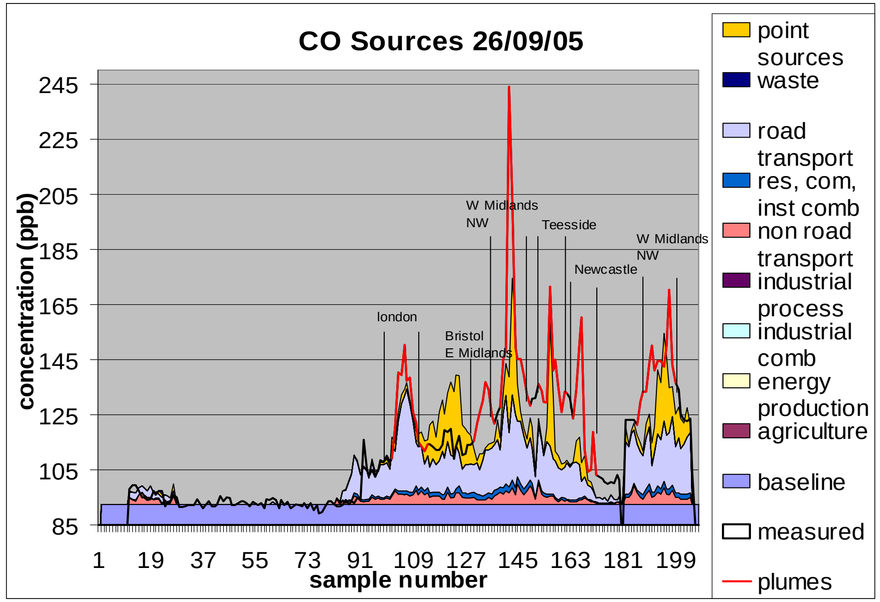


UK Facility Airborne Atmospheric Measurement (FAAM): BAe-146

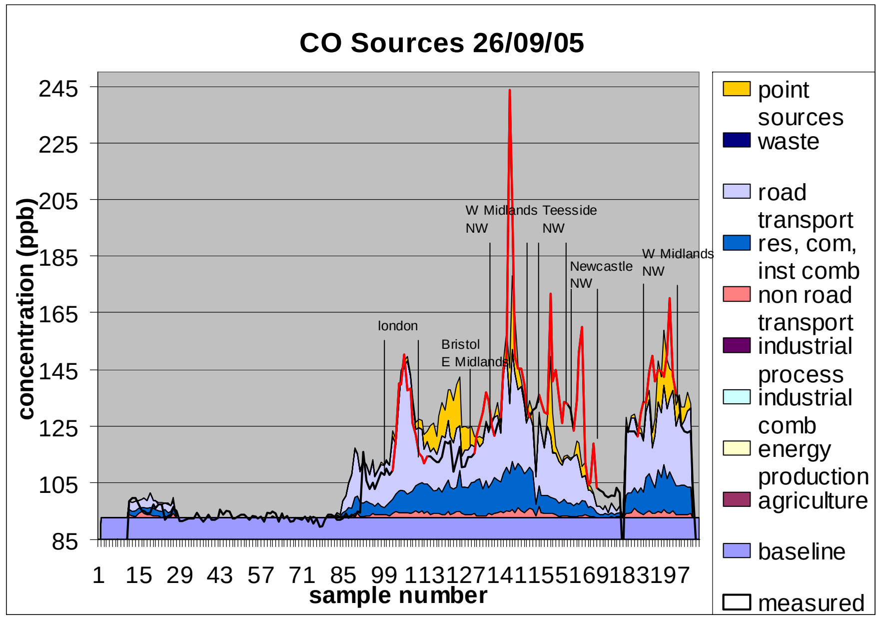
Flight duration: max. 5:30 hours



Original emissions



Adjusted emissions



Results: NAME Sectorial Emission Adjustment Technique

	CO (kT/y)	CO_2 (kT/y)	N_2O (kT/y)	CH_4 (kT/y)
NAEI (2003)	2757	572196	130	1933
B92	1900 ± 147	490000 ± 68500	880 ± 456	*8700 $\pm^{7770}_{6870}$
B97	3700 ± 605	470000 ± 117000	540 ± 427	4600 ± 1190
B102	2700 ± 250	410000 ± 93000	620 ± 334	*****
B111	*3000 ± 202	*1200000 ± 181000	*930 ± 223	*****
B112	1700 ± 292	620000 $\pm^{222000}_{130000}$	*2200 $\pm^{1260}_{239}$	9600 ± 3440
B113	2800 ± 847	840000 ± 319888	*2900 ± 893	12000 ± 6910
B118	800 ± 141	650000 ± 87266	320 ± 199	*****
B119	1900 ± 102	510000 ± 145000	190 ± 106	*****
B126	3300 ± 66	620000 ± 130000	3800 $\pm^{3760}_{3938}$	9300 ± 2730
B130*	2400 ± 48	420000 ± 82800	200 ± 72	1900 $\pm^{661}_{231}$
B132*	1600 ± 43	330000 ± 127000	390 ± 64	1600 ± 210
B134	4700 ± 167	510000 ± 75200	320 ± 186	6500 $\pm^{4070}_{3510}$
B136	2900 ± 334	580000 ± 246000	620 ± 532	*2800 $\pm^{1570}_{1390}$
AVERAGE	2400 ± 226	560000 $\pm^{139000}_{133000}$	350 ± 208	4000 $\pm^{1400}_{1290}$