Bidirectional ammonia exchange above a mixed coniferous forest

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Objectives:

- **Measuring ammonia fluxes (1999-2001) over forest subjected to high nitrogen inputs**
- **Partitioning of ammonia net-flux over stomatal and cuticular pathway**
- **Understanding the bidirectional exchange process by applying bidirectional models**

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Forest characteristics

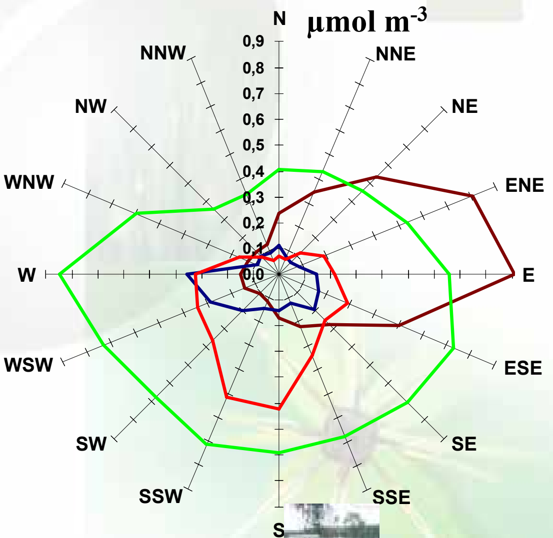
•Mixed coniferous/deciduous forest located in the Campine region

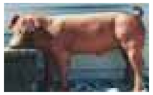
51° 18’ N, 4° 31’ E

•Forest encompasses over 300 ha

•Even-aged forest, even canopy height

•Situated between Antwerp port and agricultural area to NE





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Location measuring tower

- Scots pine stand (planting date 1929) with average canopy height of 21 m
- Carboeurope IP site and observation level II plot ICP-forest
- Nitrogen saturated forest stand:
    - Average DIN throughfall 92-2005: 35 kg N ha^{-1} yr^{-1} (70% NH_x)
    - Average NO_3 leaching 92-2005: 25 kg N ha^{-1} yr^{-1}
    - Upward NOx fluxes
    - N levels of half year’s and older needles : 2-2.5 %
    - N storage in soil and forest floor > 6000 kg N ha^{-1}
    - Annual N increment in forest floor amounts to 20 kg ha^{-1} yr^{-1}

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Measuring tower

- welded scaffolding tower of 40 m with 9 m^2 ground area
- Sonic anemometer: Gill Solent 1012 R2 20.8 Hz
- Profiles (24, 32, 40m) :
  - NH_3: AMANDA rotating wet denuder (Wyers et al., 1993): June 1999- Nov 2001
  - SO_2, NO_x, O_3, CO_2 (from 1995 onwards)
  - Wind speed (Didcot DWR-205G, UK)

- meteorological variables:
  - relative humidity and temperature (psychrometer, Didcot DTS-5A, UK)
  - leaf wetness (237F sensor grid, Campbell, UK)
  - Precipitation (tipping bucket, Didcot DRG-51, UK).
  - down-welling shortwave radiation (pyranometer, Kipp and Zonen CM6B , NL)
  - Photosynthetic photon flux density (JYP 1000, JDEC, France)

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Aerodynamic gradient method:

$$F = -K \frac{\partial \chi}{\partial z}$$

Dyer and Hicks, 1970

$$K_{NH_3} = \frac{k(z-d)u_*}{\phi}$$

d = 19.2 m;
z = $\sqrt{z_1 z_3}$ = 29.9 m;
Z_0 = 1.4 m

$$\phi_h = \begin{cases} L \le 0 \cdots\cdots\cdots \alpha * \left(1 - 16\frac{(z-d)}{L}\right)^{-\frac{1}{2}} \\ L > 0 \cdots\cdots\cdots \alpha + 5\frac{(z-d)}{L} \end{cases}$$

Bosveld, 1991

α = 0.87 RSL

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$$\upsilon_d(z-d) = \frac{-F}{\chi_{(z-d)}} = \frac{1}{R_t} = \frac{1}{R_a+R_b+R_c} \quad \Rightarrow \quad R_c = \frac{1}{\nu_d} - R_a - R_b$$

$$R_a(z-d) = \frac{1}{ku_*}\left[\ln\left[\frac{z-d}{z_0}\right] - \Psi_h\left(\frac{z-d}{L}\right) + \Psi_h\left(\frac{z_0}{L}\right)\right]$$

Garland, 1978

$$R_b = \frac{2}{k\,u_*}\left(\frac{Sc}{\Pr}\right)^{2/3}$$

Hicks et al., 1987

Rejection criteria:

- **Stationarity: |z/c*(dc/dt)| > 0.01 m s^{-1}**
- **Invalid flux-profile relationships: u* < 0.1 m s^{-1}**
- **Detection limit c < 0.1 m s^{-1}**
- **Outliers: ν_d > 2/R_a**

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Flux characteristics:

Net-flux	*Total average*	*Daytime*	*Nighttime*
n	8824	4734	4090
$[NH_3]$ ($\mu g\ m^{-3}$)	4.1 ± 6.5	4.2 ± 5.4	4.0 ± 7.5
Flux ($\mu g\ m^{-2}\ s^{-1}$)	-0.091 ± 0.176	-0.125 ± 0.222	-0.053 ± 0.085
ν_d ($cm\ s^{-1}$)	3.0 ± 4.6	3.5±5.1	2.4 ± 3.9

Daytime defined when solar radiation > 5 W m^{-2}

- 14 % emissions with ν_d of -2.1 cm s^{-1}, mostly occurring during daytime
- high variability:
  - Daytime/nighttime
  - Wind direction
  - Canopy wetness:

No rain/LW = 0:	dry canopy
No rain/ 0< LW < 1:	wet canopy
No rain/ LW = 1:	water-saturated canopy
Rain:	rainy

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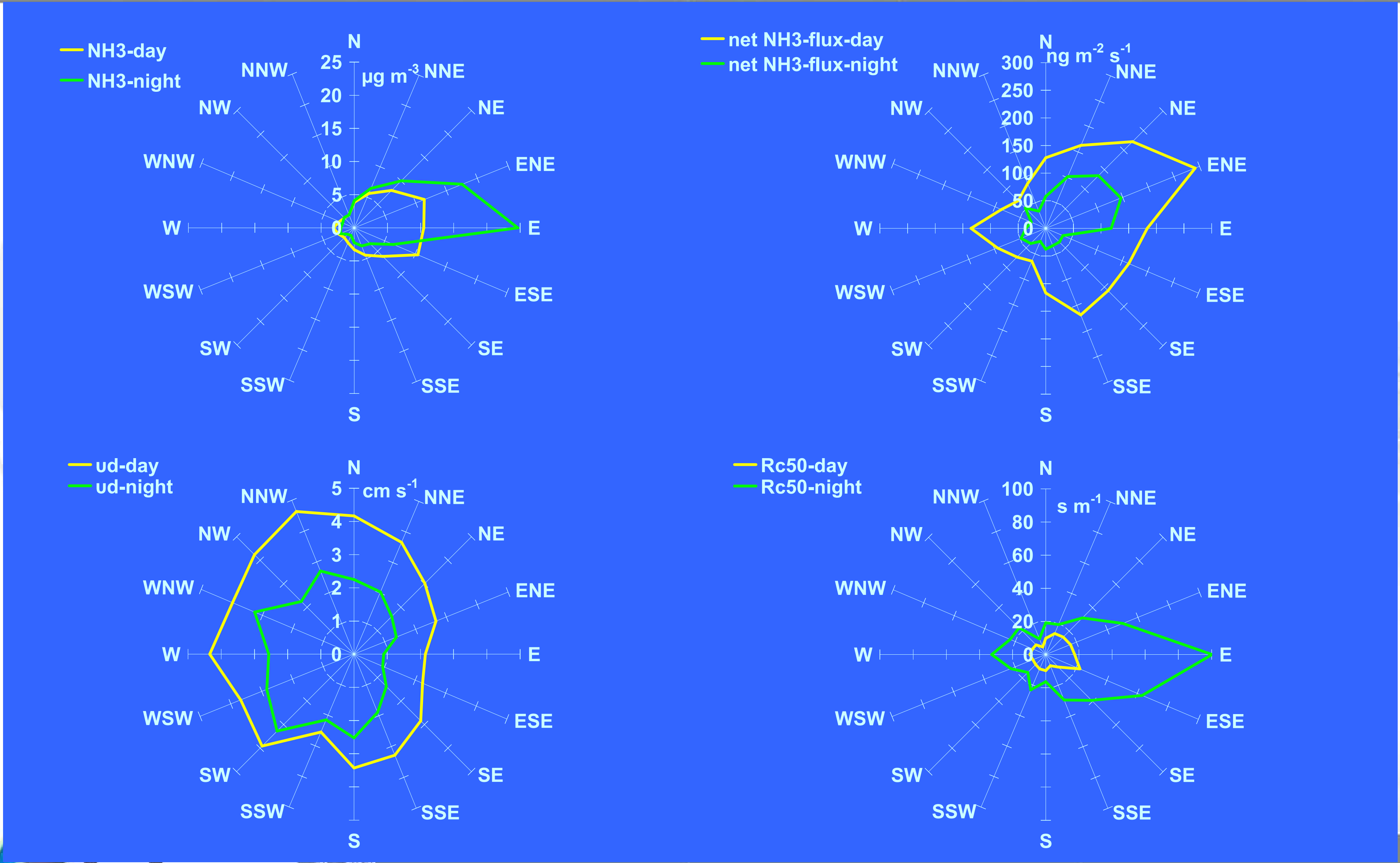

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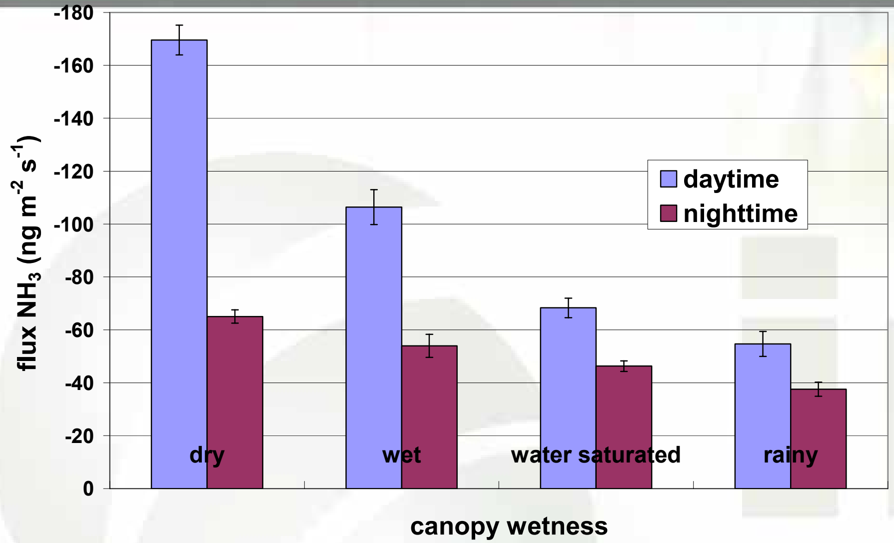

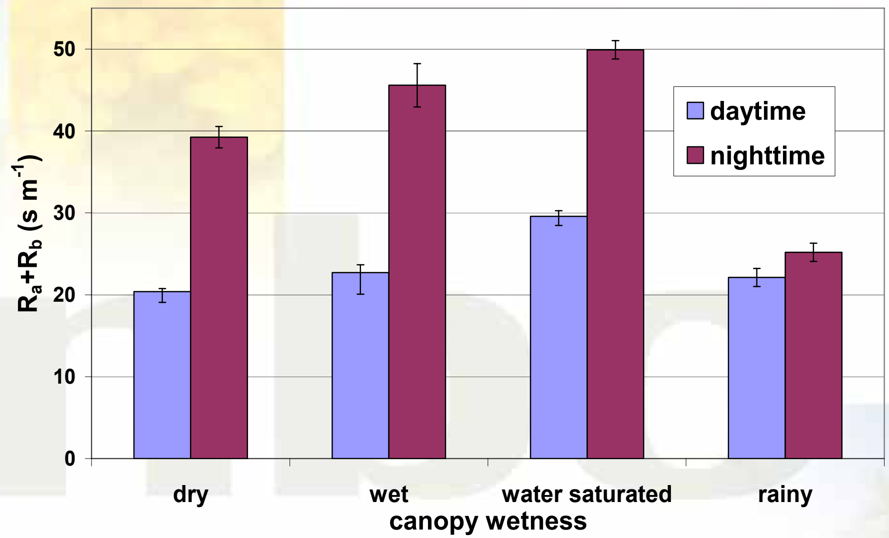

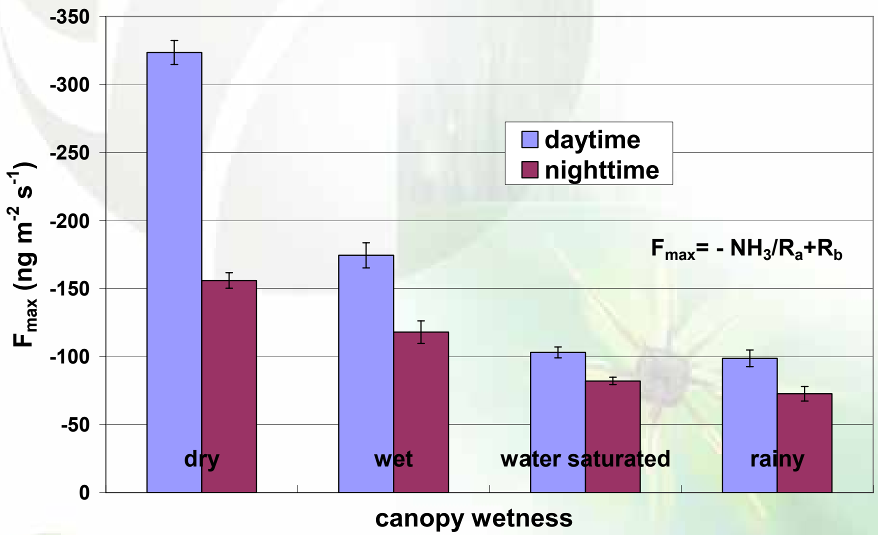

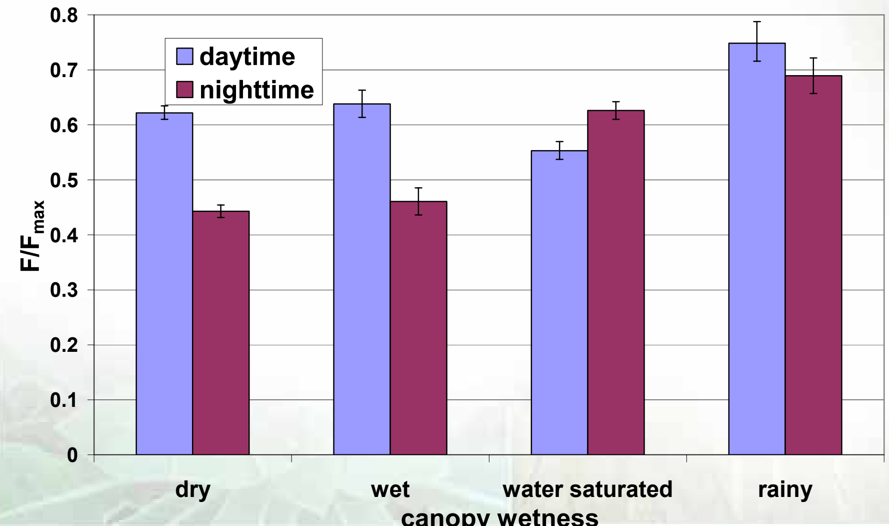

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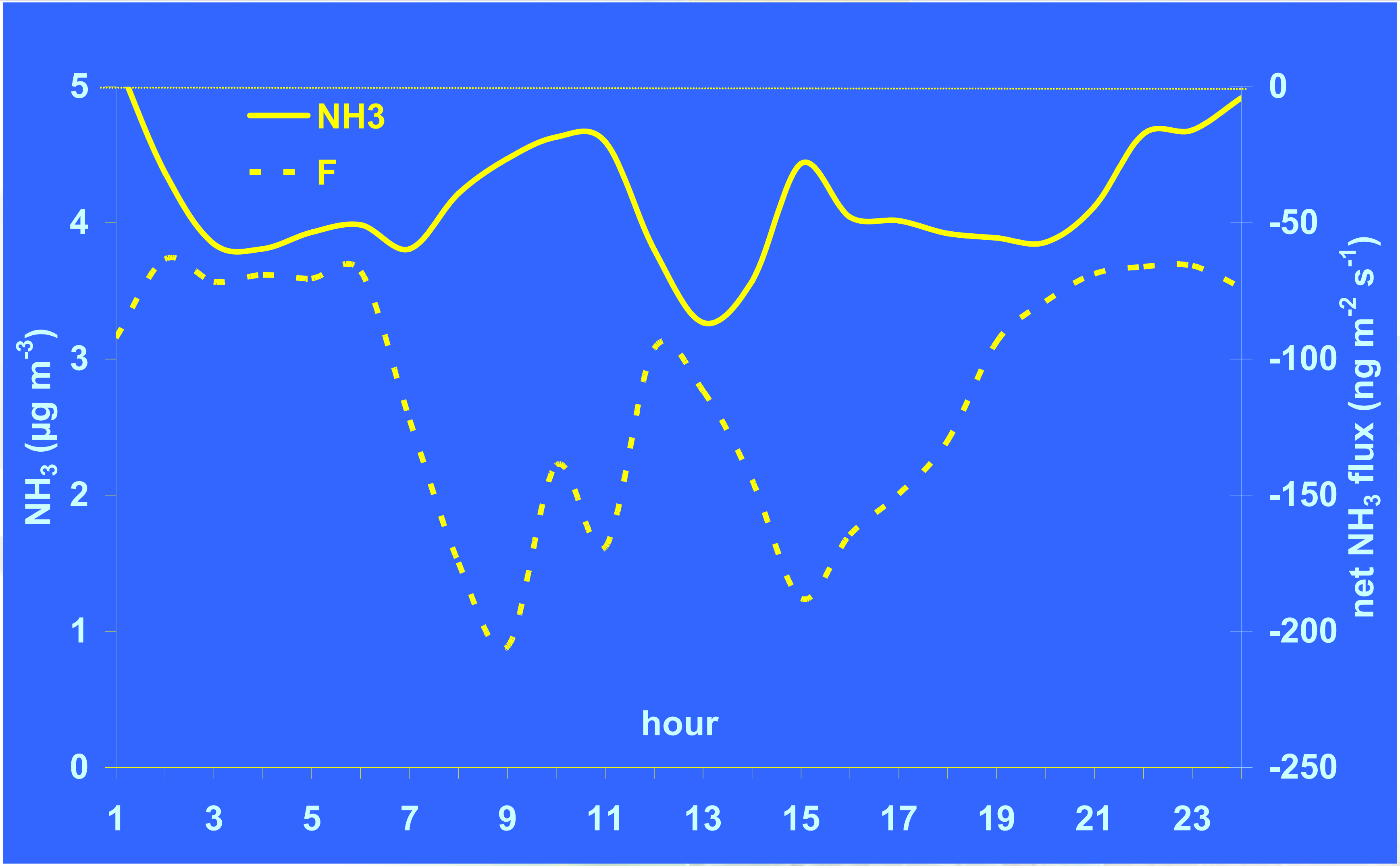

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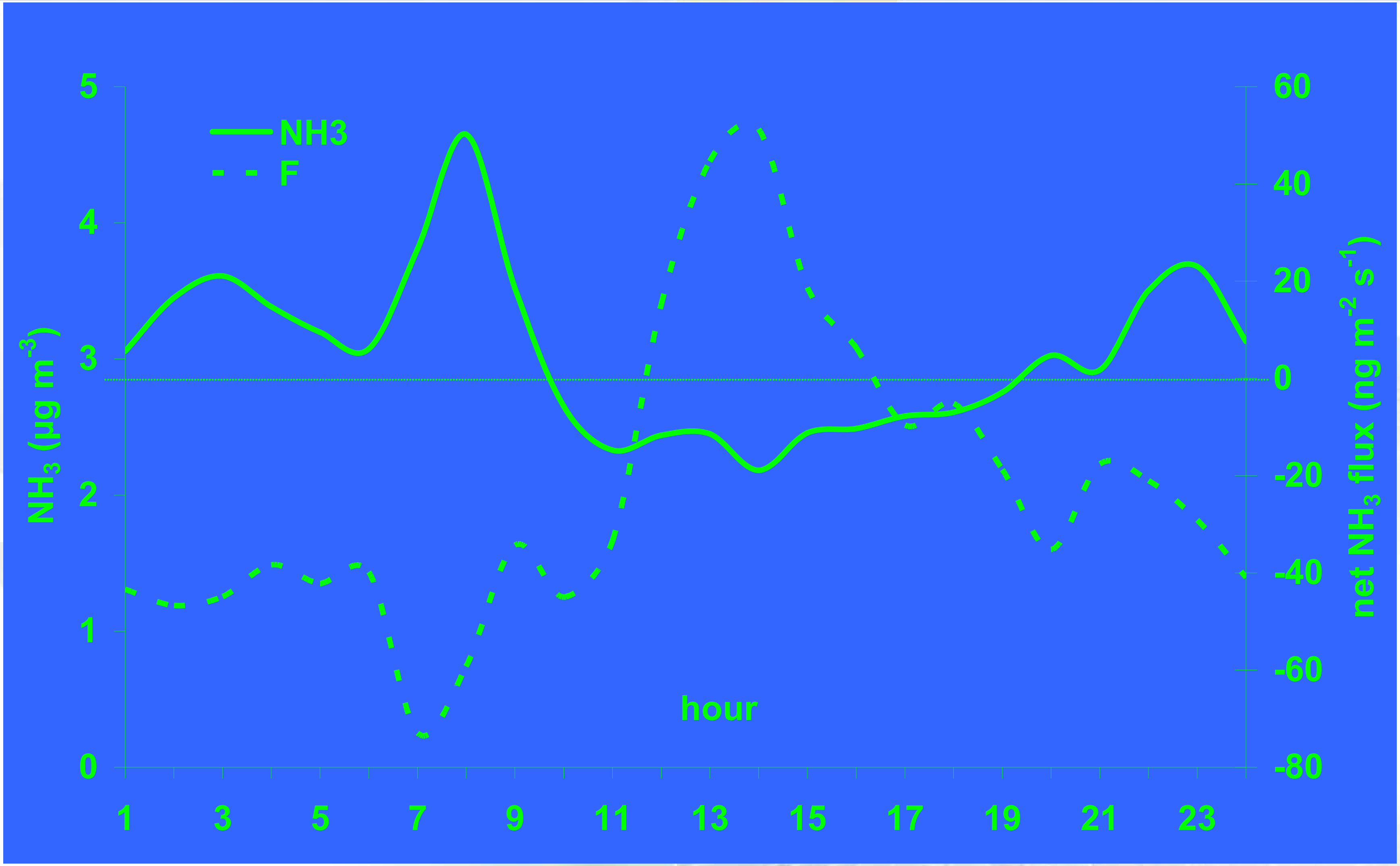

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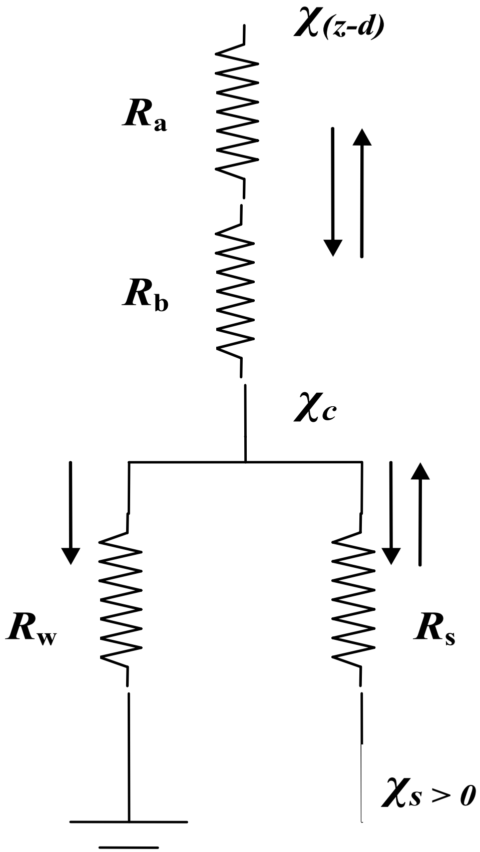
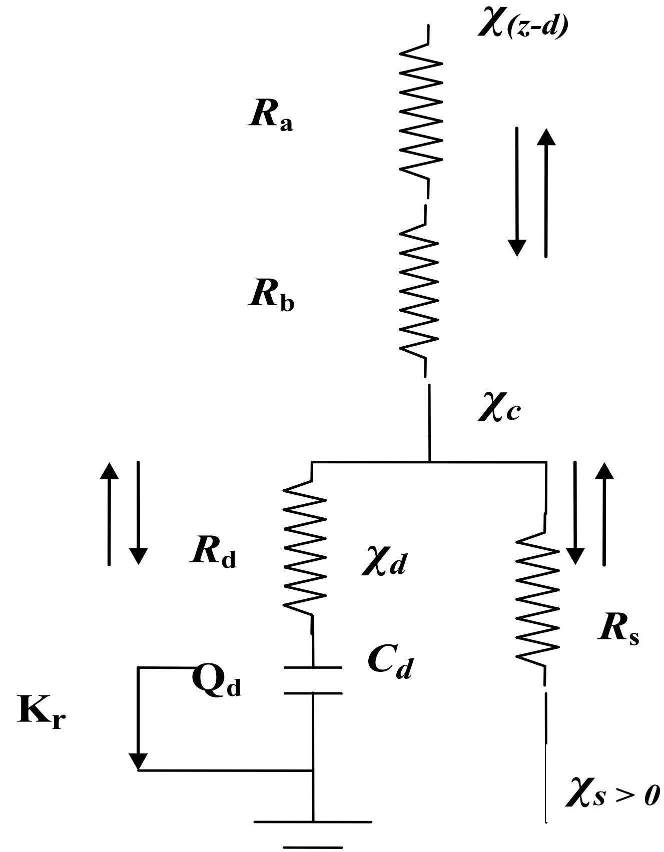
(χ_c-R_w) model (Sutton et al. ,1993)



(χ_c-C_d) model (Sutton et al. ,1998)



$$F_t = -\frac{\chi_{(z-d)} - \chi_c}{R_a\{z-d\} + R_b}$$

$$\chi_c = \frac{\left[\chi\{z-d\}/\left(R_a\{z-d\}+R_b\right)+\chi_s/R_s\right]}{\left[\left(Ra\{z-d\}+Rb\right)^{-1}+R_s^{-1}+R_w^{-1}\right]}$$

$$\chi_c = \frac{\left[\chi\{z-d\}/\left(R_a\{z-d\}+R_b\right)+\chi_s/R_s+\chi_d/R_d\right]}{\left[\left(Ra\{z-d\}+Rb\right)^{-1}+R_s^{-1}+R_d^{-1}\right]}$$

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Static Canopy compensation point-cuticular resistance (χ_c-R_w) model

$$F_t = F_s + F_w$$

$$F_s = \frac{(\chi_s - \chi_c)}{R_s}; \qquad F_w = \frac{-\chi_c}{R_w}$$

- R_s multiplicative model (*ppfd*, *vpd*, *ψ*, *T*)
- χ_s = stomatal compensation point (µg m^{-3})

$$\chi_s = \frac{161500}{T}\exp(-10380T^{-1})\frac{[NH_4^+]}{[H^+]}$$

(Nemitz et al.,2000)

- R_w parameterisations using night time R_c
  $R_w = R_{w,min}$ exp((1-RH)/a)
  - For different canopy wetness categories
  - stratified according to T (cold, warm)
  - According to molar NH_3/SO_2 ratio

(i) molar ratio < 1: excess of SO_2 over NH_3
(ii) molar ratio between 1 and 5: near-equivalent concentrations
(iii) molar ratio > 5: excess of ammonia over SO_2.

Dynamic Canopy compensation point-cuticular capacitance (χ_c-C_d) model

$$F_t = F_s + F_d$$

$$F_s = \frac{(\chi_s - \chi_c)}{R_s} \qquad F_d = \frac{(\chi_d - \chi_c)}{R_d}$$

- χ_d = Adsorption concentration (µg m^{-3})

$$\chi d = \frac{Qd}{Cd}$$

$$Cd = M_{H2O}^{c}\left[[H+]/\left(10^{1.6035-4207.6/T}\right)+\left(10^{1477.7/T-1.6937}\right)\right]$$

(Sutton et al., 1998)

- *Canopy area water film thickness* M_{cH2O} = LAI.M_{H2O}
- M_{H2O} = 0.0031.exp((3.5061*leaf wetness)
- $R_d = 5000/C_d$
- pH water film?
- *Q*d{i+t} = *Q*d{i} – *F*d.t
  *Q*d{i+t} = *Q*d{i} – *F*d.t – *Q*d{i}.K_r

K_r = adsorbed NH_4^+ reaction rate ?

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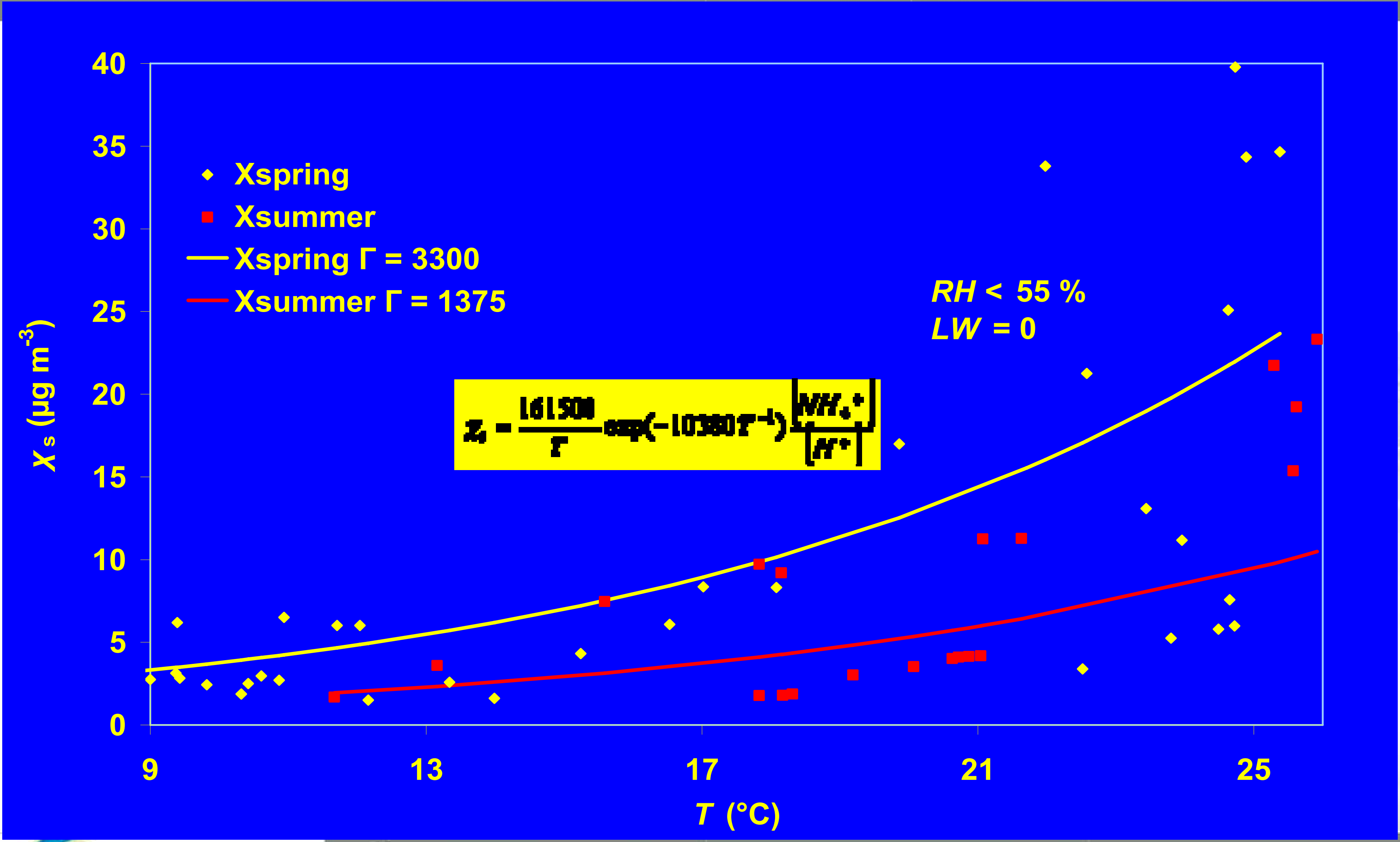

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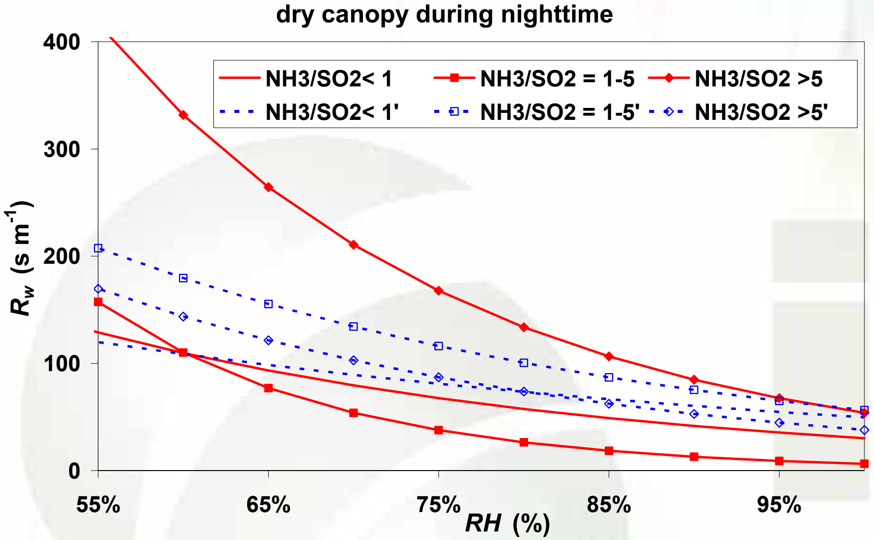

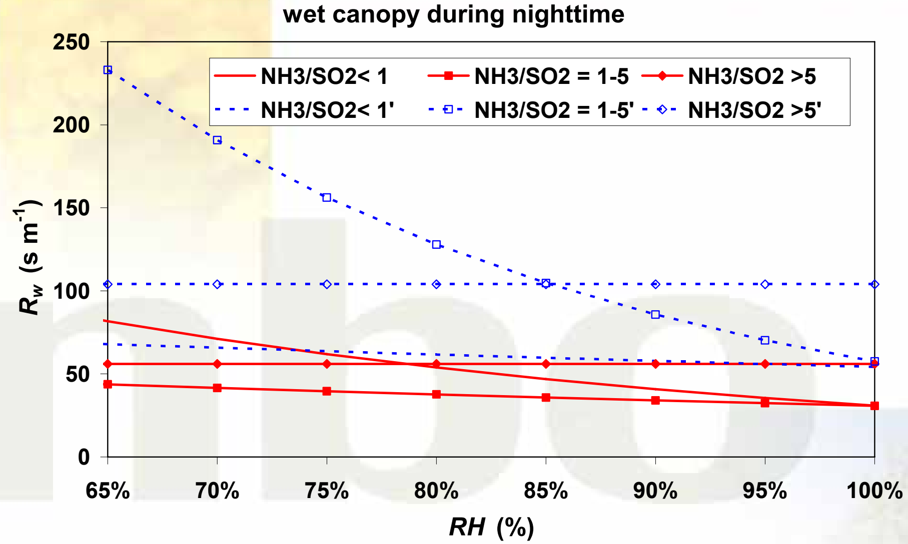

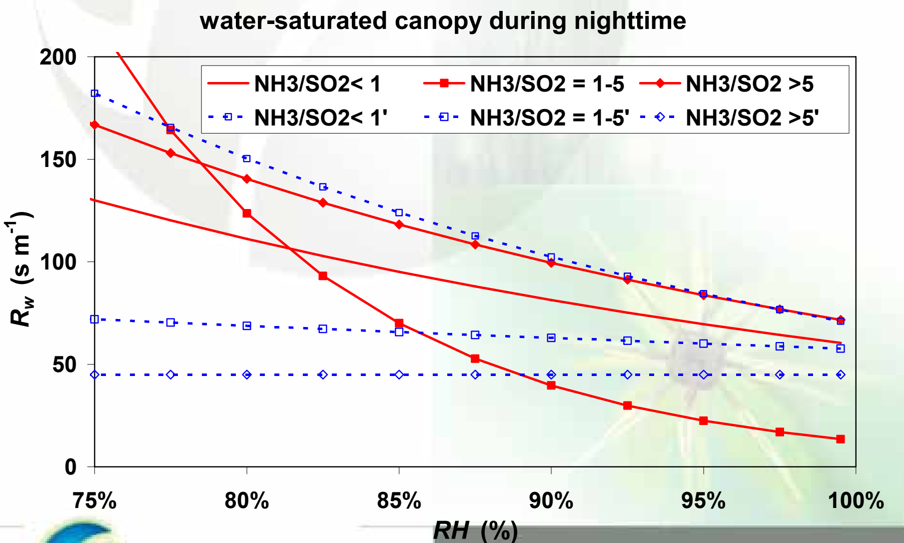

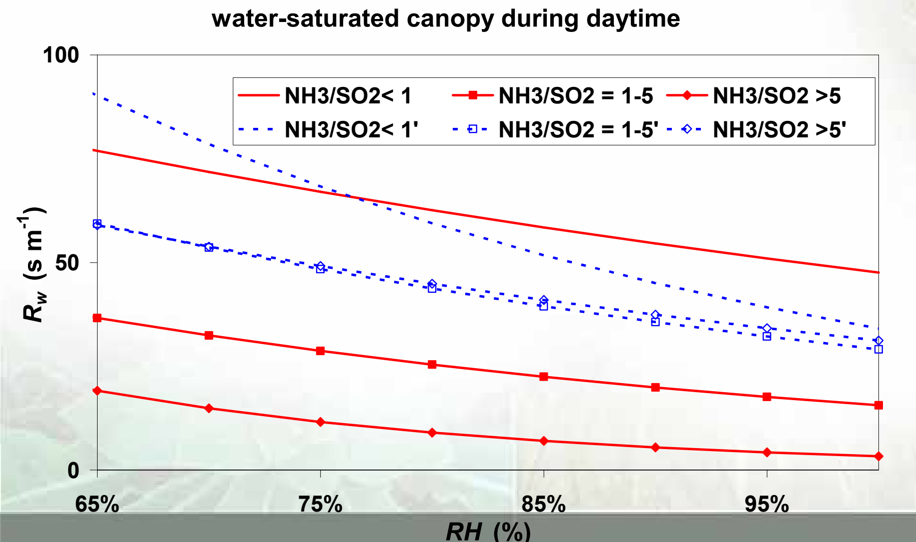

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Table: Measured versus modelled fluxes of ammonia subdivided into day- and nighttime (in ng m^{-2} s^{-1}) (modelled fluxes in bold indicate bias with measured flux > 25 %)

	Measured flux				Static model				Dynamic model			
period	n	day	night	all	day	night	all	R_2all	day	night	all	R_2all
Jun99	310	-232	-96	-186	**-76**	**-42**	**-64**	*0.80*	-216	-122	-185	*0.88*
Jul99	104	-344	-142	-270	**-256**	**-66**	**-186**	*0.69*	-314	**-232**	-284	*0.58*
Nov99	340	-26	-58	-47	**-91**	-55	**-68**	*0.03*	**-16**	-71	-52	*0.24*
Feb00	415	-41	-28	-33	**-15**	**-9**	**-11**	*0.24*	-40	-31	-34	*0.25*
Mei00	178	-211	-152	-187	-193	**-73**	-144	*0.37*	-225	-142	-191	*0.08*
Jul00	107	-69	-26	-58	**-21**	**-16**	**-20**	*0.56*	-69	-32	-60	*0.67*
Aug00	102	-73	-9	-54	**-7**	-11	**-8**	*0.82*	-65	**-19**	-52	*0.94*
Nov00	221	0	-6	-4	**-10**	-7	**-8**	*0.01*	**-2**	-6	-4	*0.30*
Apr01	109	-37	-91	-57	**-135**	**-49**	**-103**	*0.11*	-37	-90	-57	*0.29*
Mei01	169	-108	-60	-90	-94	**-35**	-72	*0.44*	-99	**-219**	**-143**	*0.35*
Aug01	253	2	-3	1	**25**	-5	**17**	*0.15*	2	-3	1	*0.27*
Sep01A	224	-42	-40	-41	-45	-49	-47	*0.31*	**-55**	**-91**	**-74**	*0.16*
Sep01B	191	7	-13	-3	**-9**	-12	**-10**	*0.10*	**12**	-17	-3	*0.40*
Okt01	369	-153	-90	-117	**-23**	**-24**	**-24**	*0.47*	-145	-90	-114	*0.91*
Nov01A	116	-2	-127	-70	**-71**	**-58**	-64	*0.11*	**-14**	-113	-68	*0.51*
nov01B	245	-15	-16	-15	-13	**-8**	**-10**	*0.36*	-18	-14	-15	*0.49*

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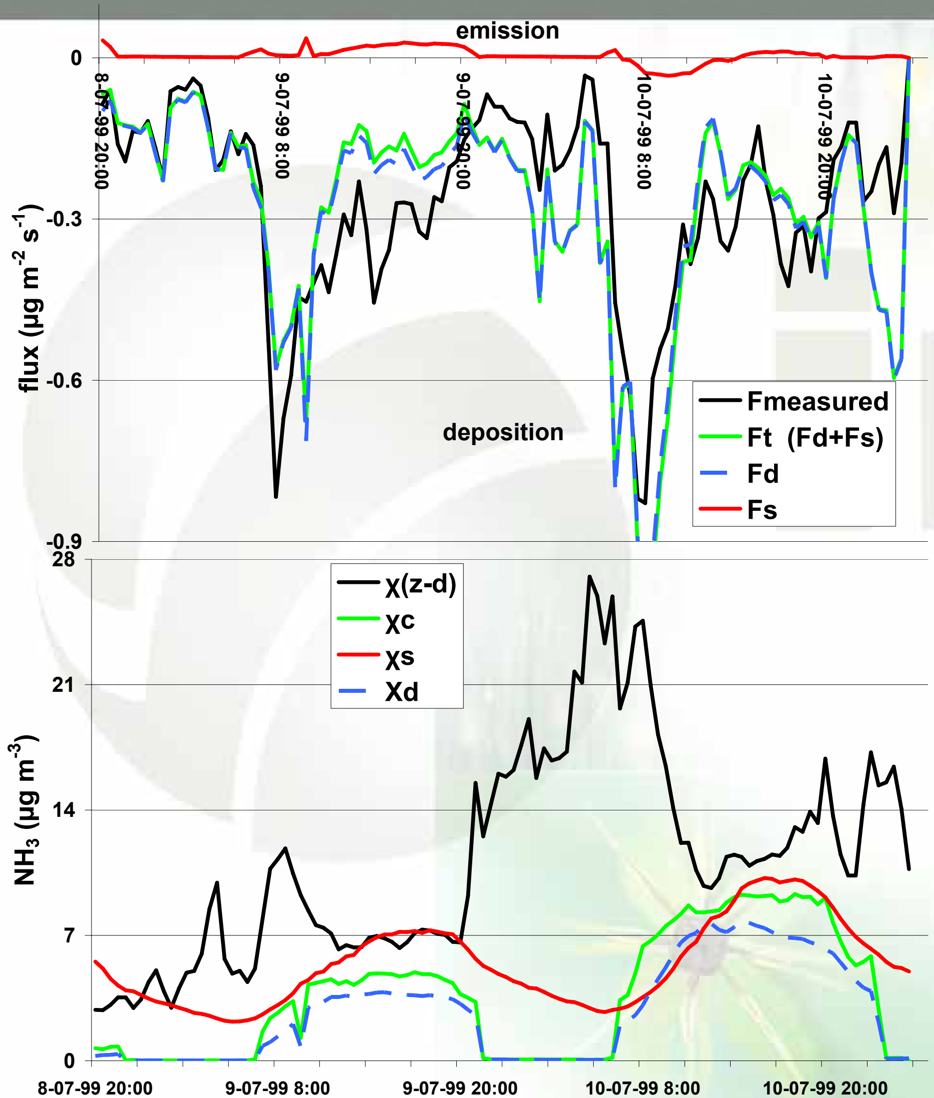

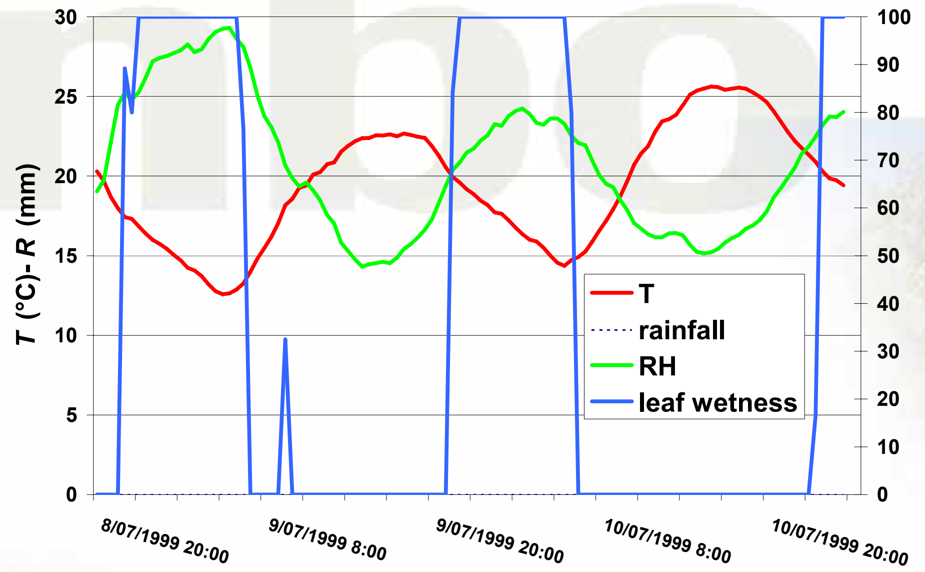

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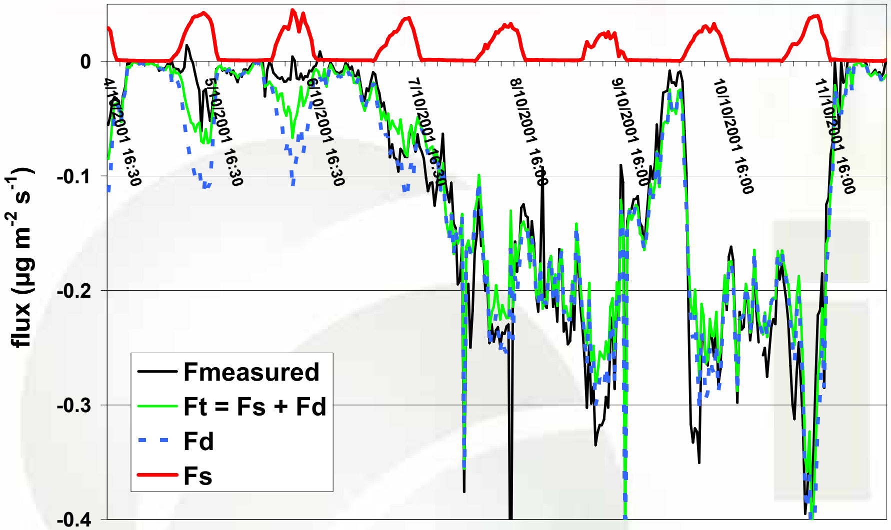

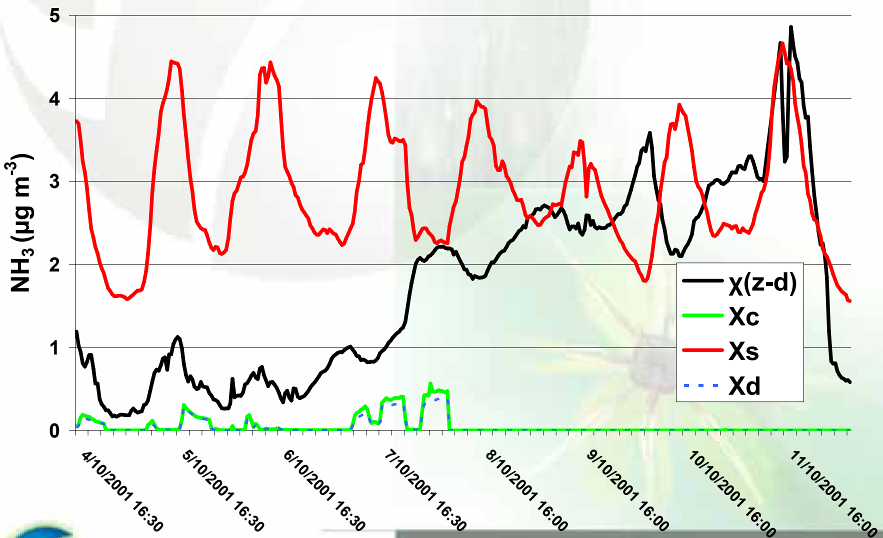

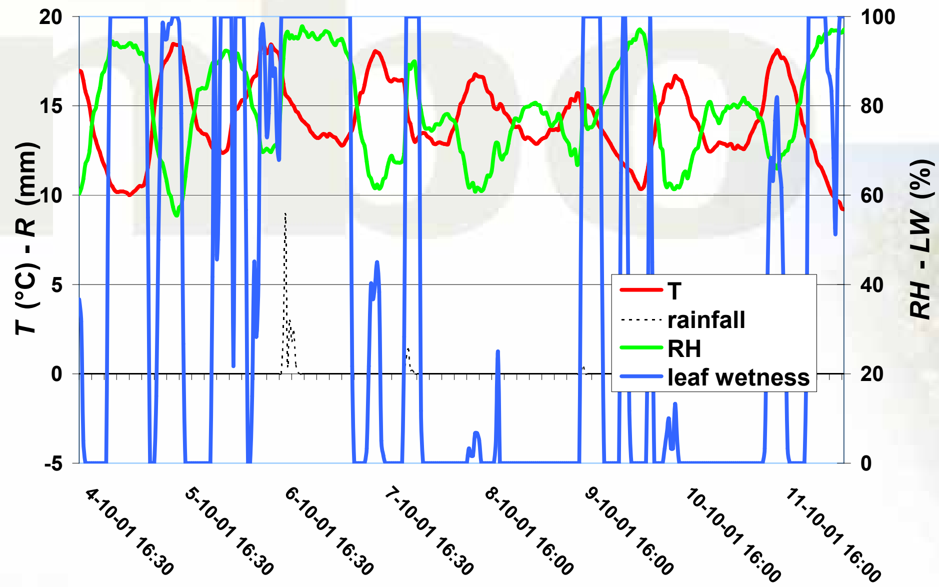

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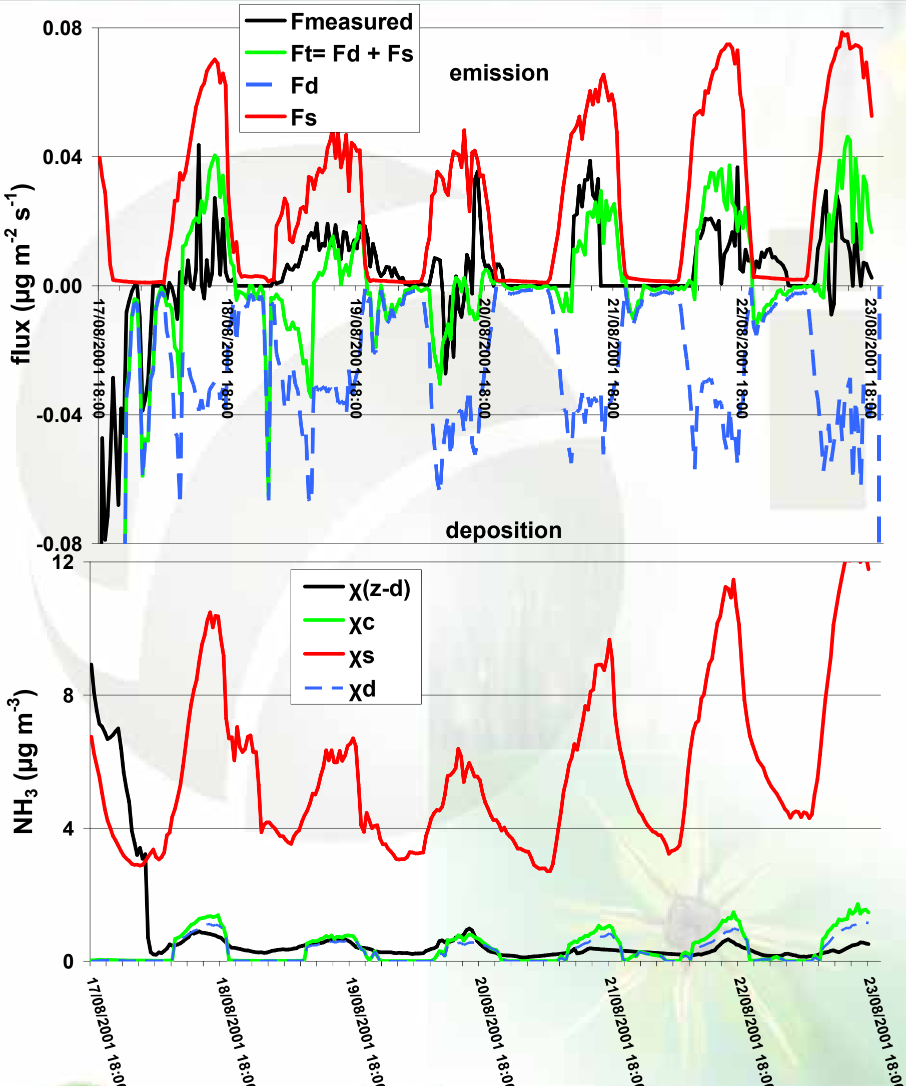

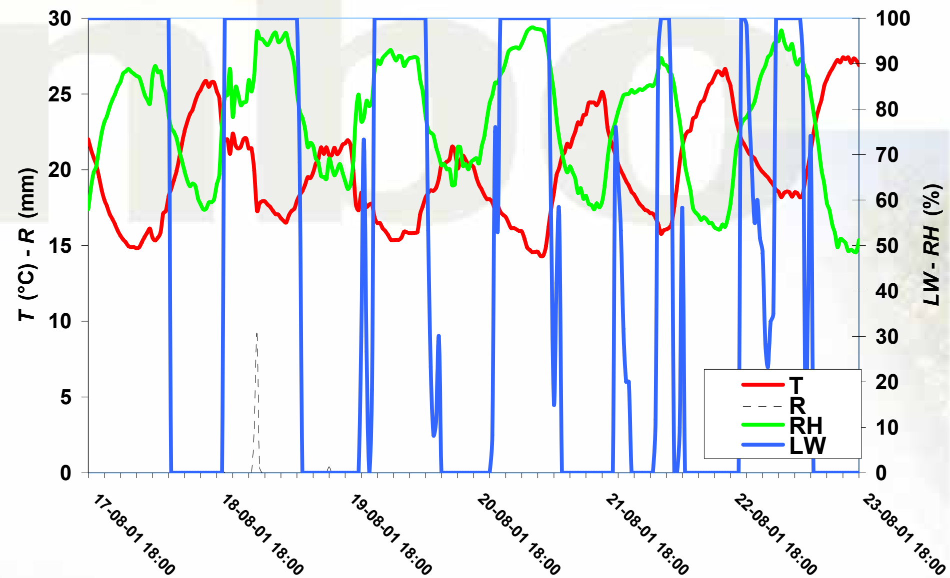

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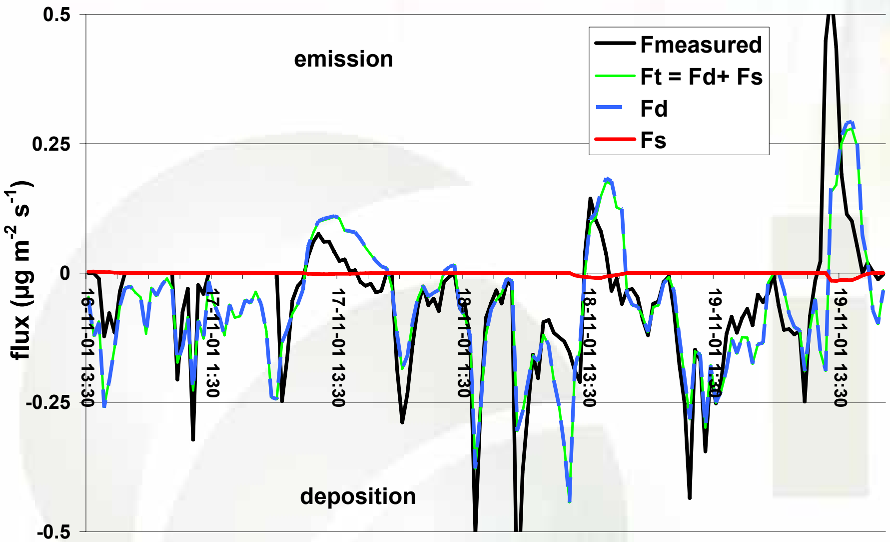

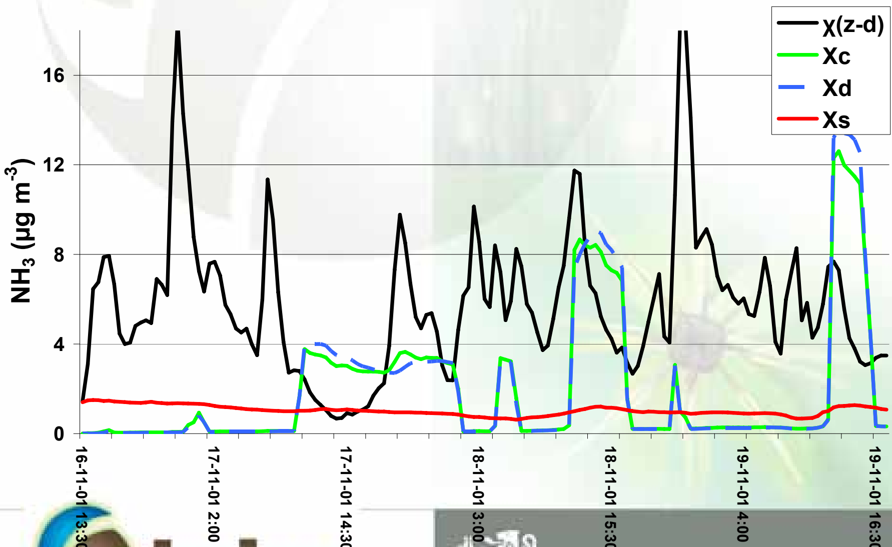

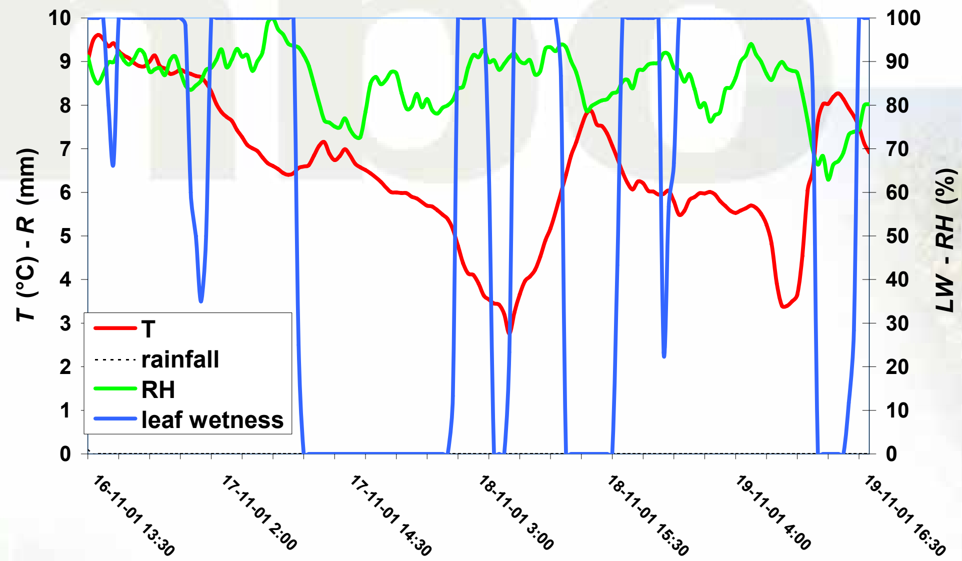

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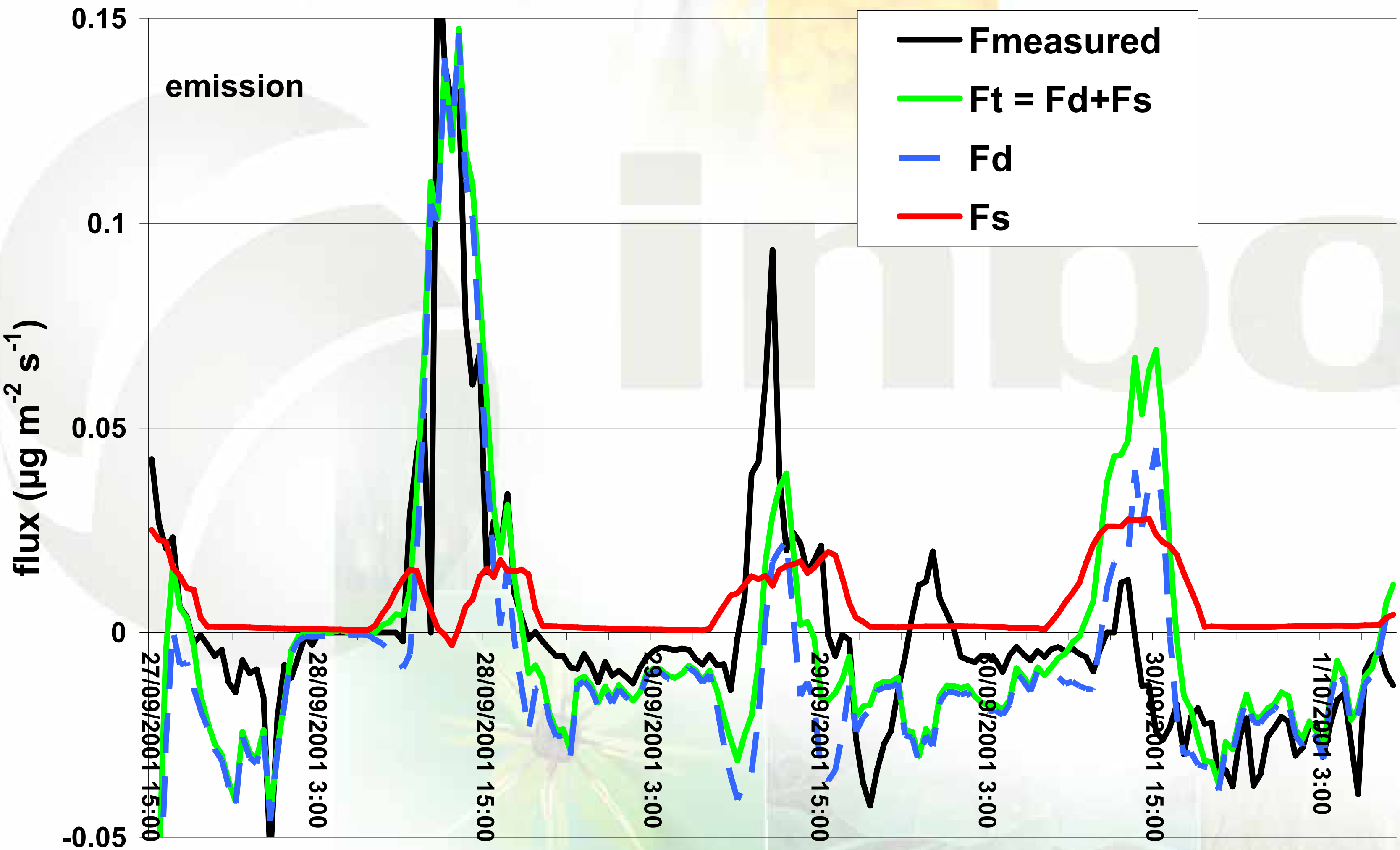

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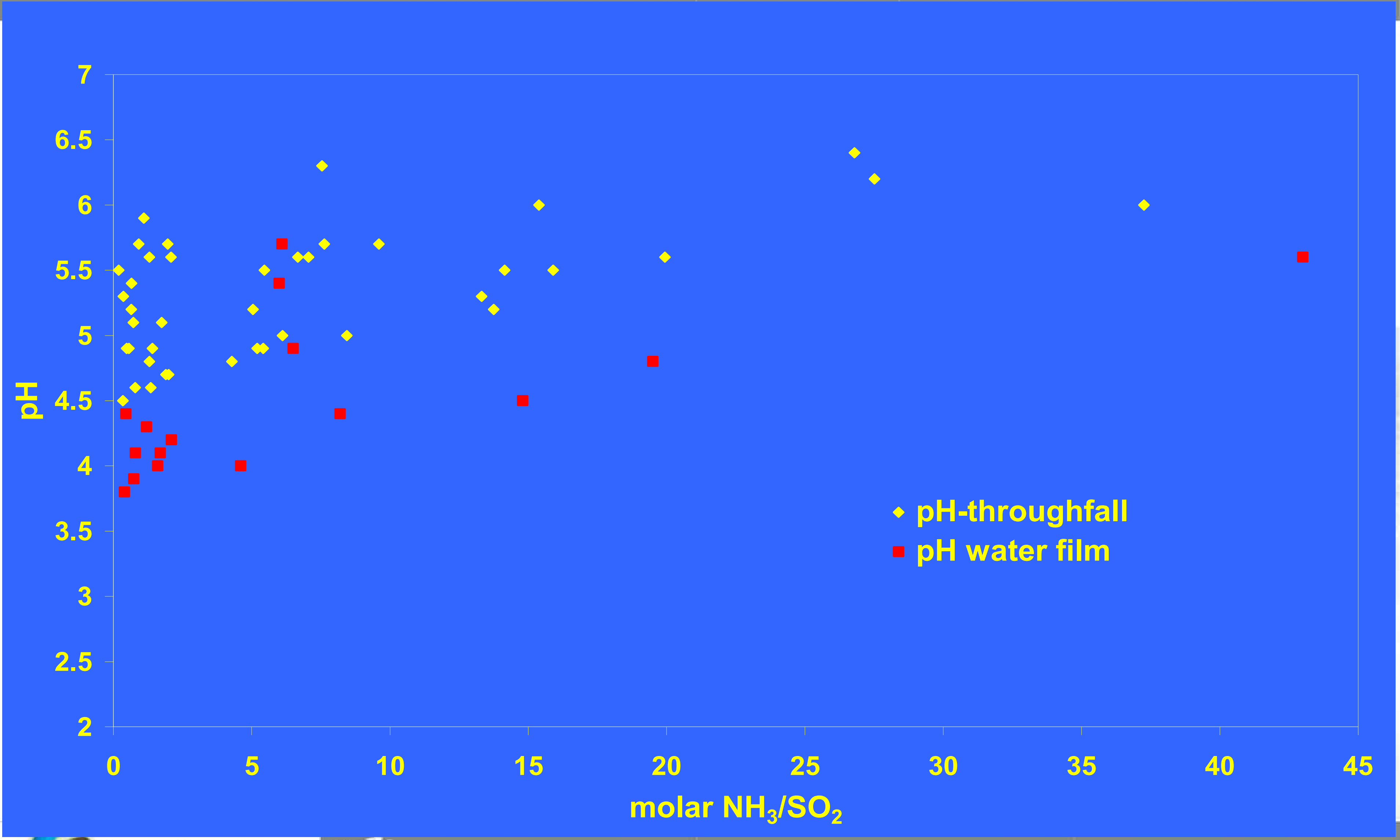

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Conclusions:

- Cuticular pathway > stomatal pathway
- Importance of canopy wetness measurements
- Both cuticular desorptions as stomatal emissions occur
- Focus:
  - Water films (thickness, acidity)
  - Deposition/emission history
  - Variability in χ_s