SURFACE-ATMOSPHERE EXCHANGE OF PARTICULATE AND GASEOUS POLLUTANTS AT THE LOCAL SCALE

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Soutenance Habilitation à Diriger les Recherches. 29 Novembre 2011.
Outline

- Context, issues and research questions
- Methodology
  - Approach
  - Models developed
  - Experimental setups
- A few key results
  - Emissions and deposition of NH$_3$ near hot spots
  - Pollen dispersal
  - Sources and sinks of pollutants and GHG in croplands
- Conclusions
- Future researches
Context - General

- Agriculture & the environment

<table>
<thead>
<tr>
<th>Environment Impact</th>
<th>GHG</th>
<th>Groundwater</th>
</tr>
</thead>
<tbody>
<tr>
<td>Global warming</td>
<td>N2O, CH4</td>
<td></td>
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<tr>
<td>N2O, CH4</td>
<td>CO2</td>
<td></td>
</tr>
<tr>
<td>Air pollution</td>
<td>NO2, O3</td>
<td></td>
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<tr>
<td>Biodiversity</td>
<td>Nr</td>
<td>GMO</td>
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<td>Acidification</td>
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<tr>
<td>Eutrophication</td>
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<tr>
<td>Ecotoxicity</td>
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</tbody>
</table>

- Pesticides
  - NH3, NO
  - NO2, O3

- Soil organic matter
- NO3
- Pesticides
Context-GHG

- Agriculture: a key in global warming?

- 10 to 12% of GHG
- Main emitter of N₂O (54%)
- Contributes to CH₄
- Terrestrial ecosystems: contributor to uncertainties:
  - Ozone budget
  - Aerosol (NH₃)
  - Future land uses
Context-Nr

The Nitrogen cascade

- Atmospheric N₂ fixed to reactive nitrogen (N₄)
- Nitrous Oxide (N₂O)
- Nitrogen oxides (NOₓ)
- Ammonium nitrate in rain (NH₄NO₃)
-Further emission of NOₓ & N₂O carrying on the cascade
- Terrestrial eutrophication
- Soil acidification
- Leached nitrate (NO₃⁻)
- Water eutrophication
- Nitrate in Streamwaters

(Tg N y⁻¹ EU27 in 2000)
Context-GMO

- World production of GMO increasing
  - Gene transfer (oilseed rape),
  - pesticides
  - quality

GLOBAL AREA OF BIOTECH CROPS IN THE WORLD
Million Hectares (1996-2010)

(ISAAA, 2010)
Issues-GHG

Can we lower GHG emissions?

Carbon loss
0.9 to 2.2 Mg C ha\(^{-1}\) y\(^{-1}\)
Uncertainty > 100%

Issues

- Can we lower GHG emissions?

Carbon loss

Deposition:
40% of tropospheric ozone budget

\[ \text{O}_3 \text{ deposition} \]

- Carbon assimilation
- \( \text{O}_3 \) balance

\[ \text{C, N and GHG budgets of crops?} \]
- Evaluate management

Field scale
Long term
Issues-Nr

- Can we reduce NH$_3$ emissions?

Emissions
97% from agriculture

Short range
Process studies
Mitigation

Local
Recapture?

Indirect
emissions

Reduction
methods

Long range
transport

FIELD SCALE
SHORT-RANGE
LONG-RANGE
Issues-GMO

- Can we control gene flows?

- pollen emission process

- Potential gene transfer

  - Short range
  - Short-range process studies
  - Mitigations
  - Maize

- Long range

FIELD SCALE
SHORT-RANGE
LONG-RANGE
Research questions

(1) Quantify
(2) Understand processes
(3) Evaluate mitigation options

Sources and sinks in agrosystem

CO$_2$  O$_3$  NO$_x$

Emissions

NH$_3$

Pollen

Particles

Pesticides

Short range deposition

FIELD SCALE

SHORT-RANGE

C & N balances
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Methodology-1

- Operational models
- Process modelling
- Controlled conditions
- Dissemination
  - Feedback

- Field experiments
- Parameterise
- Question
- Validate
- Calibrate
Methodology-1

- Eddy covariance
- Gradients
- Mass balance

Dispersion models
- Resistance models
- Inverse models

Dispersion
Surface exchange

Chambers
Apoplast extraction

Dissemination

feedback
Methodology-2

- Lagrangian Stochastic dispersion and deposition models

- Gases (MODDAAS)  
- Particles (SMOP)
Methodology-2

Concepts in MODDAAS

\[ C_i = \sum_{all\_sources} D_{ij} S_j (C_j) \]

- \( S \): sources (negative = sink)
- \( D_{ij} \): dispersion matrix
- \( C \): concentration

\( D_{ij} \) is determined with the LS model.

These two schemes are equivalent

Coupling with a \( k-\varepsilon \) turbulence model (Thetis)

(Hensen et al., 2009; Loubet, 2000; Loubet et al., 2009a; Loubet et al., 2003; Loubet et al., 2006; Loubet et al., 2010; Loubet et al., 2009b; Loubet et al., 2001; Nemitz et al., 2009; Sutton et al., 2009a; Sutton et al., 2009b)
Methodology-2

■ Concepts in SMOP

■ Particle is “Settling while travelling”

\[ \Delta z = (w' + \bar{W} - V_s) \Delta t \]

■ Cross-trajectory effect reduces the “Particle” Lagrangian time scale

\[ T_L^P = \frac{T_L}{\sqrt{1 + \left( \frac{\beta \cdot V_s}{\sigma_w} \right)^2}} \]

■ Deposition is statistically treated

(Bouvet et al., 2007; Jarosz et al., 2005; Jarosz et al., 2003; Jarosz et al., 2004)
Methodology-3

- ROSAA analyser: NH$_3$ fluxes

Methodology-3

Settling speed of pollens

(Jarosz, 2004; Loubet et al., 2007; Marceau, 2011; Marceau et al. 2011a, 2011b)
Methodology-4

- FIDES Inverse modelling: combining modelling and measurements

Inversion: finding $C_c$ knowing $C(z_{\text{ref}})$

$$C(x, z) = \frac{Q \cdot A}{x^\beta} \exp \left[ - \frac{Z^\alpha + Z_s^\alpha}{c \cdot x} \right]$$

$$Q(x) = - \frac{C(x, z_s) - C_c}{R_b}$$

(Loubet et al. 2001, 2010; Hensen et al. 2009; Carozzi et al., in prep)
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Emission and deposition near NH₃ hot spots

- NH₃ losses ~ 37% of N-NH₃ applied

Other datasets
- ROSAA gradient validated
- FIDES inverse methods valid (~10% error)
- FIDES Integration over monthly periods (< 30% error)

Deposition of NH$_3$ near hot spots

Short-range deposition mostly sensitive to
- The wind speed and release height
- The surface resistances (cuticular and stomatal)
- The ammonia potential emissions of surrounded fields

(Loubet 2000; Loubet et al., 2001; 2009)
Mitigating hot spots NH$_3$ emissions

- Tree belt can recapture NH$_3$ up to
  - 30% for housing
  - 20% for lagoon
  - 60% for understorey

- Deposition is proportional to the LAI if the canopy is homogeneous

- Open trunk spaces disfavour NH$_3$ recapture

- Dense canopies at the source height should be favoured.

(Loubet, 2000; Loubet et al. 2009)
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Pollen settling velocity

- Settling velocity \( (V_s) \) is bi-modal
- Each mode has a given
  - Water content
  - Viability

\( V_s \) is a good proxy for viability determination

(Jarosz, 2005; Loubet et al., 2007; Marceau, 2011; Marceau et al. 2011)
Pollen dispersal and deposition

- Deposition velocity in the lee of the field is larger than in $V_s$

Maize pollen

- Preferential sweeping and clustering may explain this result when $St \sim 1$.
- $St = \frac{\tau_p}{\tau_k} \sim 1$
- The turbulence time scale equals the response time of the particle

(Jarosz, 2005; Jarosz et al. 2004, 2005)
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Sources and sinks of ammonia in cut and uncut grassland

- After cutting the NH$_3$ emissions increases because:
  - Temperature increases
  - Aerodynamic resistances ($R_a$, $R_{ac}$) and $R_b$ are low
  - Moisture dynamics links to efficient mineralization of organic nitrogen

(David et al., 2009; Personne et al. 2009; Nemitz et al. 2009; Sutton et al., 2009)
Drivers of the ammonia compensation point

- $\text{NH}_x$ mainly produced by photorespiration ($\text{F}_{\text{pho}}$)
- GS/GOGAT is very efficient in recycling the $\text{NH}_x$ into Amino acids
- $\text{NH}_x$ in the cytoplasm is therefore maintained low
- Cytoplasm and apoplasma are relatively isolated in terms of $\text{NH}_x$
- Root uptake was found the main driver of $\text{C}_s$
- Great sensitivity to active transport (focus for further research)

Massad et al. 2010
O₃-NO interaction near the surface

- When [NO] > ~1 ppb, the NO-O₃ reactions in the canopy could represent a large additional conductance (up to 1 cm s⁻¹)
- A simple chemical scheme could explain this additional destruction of O₃ in the canopy.

\[
\tau_{\text{trans}} \gg \tau_{\text{chem}} \\
NO + O₃ \rightarrow NO₂ + O₂ \\
NO₂ \rightarrow NO + O^• \\
r_{\text{chem}}(O₃) = \frac{\tau_{\text{chem}}(O₃)}{h_c} = \frac{1}{k_r \times [NO] \times h_c}
\]

Lamaud et al. (2009)
Carbon fluxes in a crop rotation

- Crop rotation was a **net loss** of carbon over 5 years 1.3 MgC ha\(^{-1}\) y\(^{-1}\)
- NEP of 4.4 MgC ha\(^{-1}\) y\(^{-1}\)
- Respiration the main driver of the yearly variability

Loubet et al. (2011)
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Conclusions-1
New concepts & knowledge

- Sources and sinks in an agroecosystem

- Magnitude of NH₃ sources and sinks in agroecosystems
- Conceptual NH₃ compensation point
- Parameterisation of deposition resistances
- Explicit role of microclimatic conditions
- Quantification of GHG N and CO₂ fluxes
Conclusions-1
New concepts & knowledge

- Short-range dispersion of pollens

  - Pollen release depends on microclimate
  - Deposition velocity increases in the lee of canopies
  - Maize pollen settling speed is bimodal
  - Settling speed, a proxy for pollen viability
Conclusions-2
Dissemination to the community

- Experimental
  - An analyser for measuring NH$_3$ fluxes (Patent pending)
  - A controlled flux chamber for NH$_3$ and O$_3$
  - An integrated Eddy covariance system
  - A settling tower for biotic particles
  - A long term monitoring flux site

- Modelling
  - A Lagrangian Stochastic model of NH$_3$ and pollen deposition near hot-spots
  - An operational model for emissions estimation (FIDES)
Conclusions-3

Answering the bigger questions?

- Can we lower GHG emissions?
  - C:N ratio around 20
  - "Avoiding N losses helps sequestering C"
  - Reduce uncertainty!

Loubet et al., in prep.

N leaching
N dry dep. (Vries, 2008)  
C:N = 30-70
Conclusions

Answering the bigger questions?

- Can we reduce NH$_3$ emissions?

  - NH$_3$ incorporation can be efficient
  - Recapture may be efficient
  - Understorey farming is an option
  - Better evaluation of NH$_3$ reduction methods is required

Volatilisation (% TAN)

<table>
<thead>
<tr>
<th></th>
<th>0%</th>
<th>30%</th>
<th>60%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incorporated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24h</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>48h</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Surface appl.</td>
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</tbody>
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60% reduction in emissions Understorey
Conclusions-3
Answering the bigger questions?

- Can we control gene flows?
- Always a small fraction of pollen flying away
- An operational model available for testing purpose

<table>
<thead>
<tr>
<th>Scénarios</th>
<th>Isolement de la parcelle « semences » d'une parcelle :</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>« semences »</td>
</tr>
<tr>
<td></td>
<td>« hybrides »</td>
</tr>
<tr>
<td></td>
<td>(IV) 50m</td>
</tr>
<tr>
<td></td>
<td>(V) 100m</td>
</tr>
<tr>
<td></td>
<td>(I) 100m +20m mâle</td>
</tr>
<tr>
<td></td>
<td>(II) 150m</td>
</tr>
<tr>
<td></td>
<td>(III) 200m</td>
</tr>
<tr>
<td>« vent faible »</td>
<td>0.27 %</td>
</tr>
<tr>
<td></td>
<td>0.24 %</td>
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<tr>
<td></td>
<td>0.2 %</td>
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<tr>
<td></td>
<td>0.42 %</td>
</tr>
<tr>
<td></td>
<td>0.37 %</td>
</tr>
<tr>
<td>« froid et humide »</td>
<td>0.35 %</td>
</tr>
<tr>
<td></td>
<td>0.28 %</td>
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<tr>
<td></td>
<td>0.42 %</td>
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<td></td>
<td>0.67 %</td>
</tr>
<tr>
<td></td>
<td>0.57 %</td>
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<tr>
<td>« vent fort »</td>
<td>1.75 %</td>
</tr>
<tr>
<td></td>
<td>1.3 %</td>
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<tr>
<td></td>
<td>3.78 %</td>
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<td>4.56 %</td>
</tr>
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<td>3.79 %</td>
</tr>
<tr>
<td>« froid et humide »</td>
<td>2.31 %</td>
</tr>
<tr>
<td></td>
<td>1.73 %</td>
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<tr>
<td></td>
<td>5.33 %</td>
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<td></td>
<td>6.23 %</td>
</tr>
<tr>
<td></td>
<td>5.2 %</td>
</tr>
</tbody>
</table>

Thèse Alexis Marceau (2011)

Tab. IV-2. Pourcentage moyen de pollen exogène viable dans la parcelle « semences »
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  - Example experimental setups
- Three key results
  - Emissions and deposition of NH$_3$ near hot spots
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- Future researches
Still challenges to face in agro-ecosystems

- To reduce uncertainties in C, N and GHG flux
- To work out the links between C, N, GHG, and pollutant fluxes
- To propose and evaluate innovative agricultural systems to reduce environmental impacts

Uncertainty (C: 85%) (N: 115%) (GHG:75%)
Proposed approach

- **Improve** the predictive capacity of surface-atmosphere exchange models for GHG and pollutants in agro-ecosystems

- **Quantify** GHG, pollutants and C/N balances in agro-ecosystems over the long term
(1) Improve the predictive capacity of surface-atmosphere exchange models

- Model coupling: The “Bioatm-EGC” model

- CHIMERE (I. Coll)
- NitroScape (J.L Drouet)
- CERES-EGC (R. Massad)

- SURFATM (E. Personne)
- SMOP / MODDAAS Aerosols
  - Chemical reactions
- VOLT’AIR (S. Génermont, C. Bedos)

- Nutrient uptake
- Chemistry (canopy)
- Cuticular uptake
- Test and validate

\[
\begin{align*}
\chi_{\text{surf}}(\text{NH}_3) & \quad R_w(\text{NH}_3) \\
\chi_{\text{surf}}(\text{NH}_3) & \quad R_{bi}(\text{NH}_3) \\
\chi_{\text{surf}}(\text{NH}_3) & \quad R_3(\text{Z}_{\text{ref}}) \\
\chi_{\text{surf}}(\text{NH}_3) & \quad R_{ac} \\
\end{align*}
\]
(2) Quantify GHG, pollutant and C/N balances in agro-ecosystems over the long term

- Key questions in long term studies
  - Nitrogen balance
  - Spatial heterogeneity of the fluxes
  - Non closure of energy balance
  - Gap filling methodologies

- Efforts on
  - Reactive nitrogen fluxes (NO₂, HNO₃)
  - N₂ fluxes (but how?)

- Methods
  - Operational tools NH₃ emissions (model + alpha badges)
  - Up to date methods (Eddy Covariances (EC), Disjunct EC, …)
(2) Quantify GHG, pollutant and C/N balances in agro-ecosystems over the long term

- **N balance of a crop rotation**

![Diagram showing N balance of a crop rotation](image)

- Leaching
- Spatial heterogeneity
- Surface exchange
- $N_2$ fluxes

Particles in sub-urban areas?

*Velthof et al. (2009)*
(2) Quantify GHG, pollutant and C/N balances in agro-ecosystems over the long term

INRA Grignon (crops)
INRA Lusignan (grassland)
ICOS network
**Strategy & collaborations**

- **Core collaborations**
  - Lab (fluxes, chambers, models)
  - INRA (Ephyse, Lusignan, Orléans, Nancy)
  - LISA, LSCE, IRSN
  - CESBIO, LA, ...
  - CEH (UK)
  - MPI (Mainz)

- **Prospective**
  - VOCs fluxes (LSCE, CEH)
  - Aerosols fluxes (IRSN, EPHYSE)

- **Projects**
  - ICOS (PI)
  - INGOS (PI)
  - ECLAIRE (PI)

- **Models & Meas**
  - ICOSERES-EGC, Surfatm, Volt’air
  - NitroScape
  - Chimère
  - SMOP
  - MODDAAS

- **PhDs**
  - Elise Potier
    - in-canopy ($O_3$-COV-NO$_x$) modelling

- **Prospective**
  - VOCS fluxes (LSCE, CEH)
  - Aerosols fluxes (IRSN, EPHYSE)
A collaborative work
Thanks to many people …

**PhD Students**
- Nathalie JAROSZ (now MC at CESBIO)
- Thomas BOUVET (now at ESA)
- Rea S. MASSAD (now CR in our lab)
- Alexis MARCEAU (now post-doc at ARVALIS)
- Patrick STELLA (soon post-doc in MPI Mainz)
- Elise POTIER (just started her PhD)

**International Partners**
- CEH Edinburgh (Mark A. SUTTON & co)
- ECN Petten (Arjan HENSEN, René OTJES & co)
- ART Zurich (Albrecht Neffel & co)
- CRA Bari (Gianfranco RANA, Rosanna FERRARA)
- Wuppertal University (Joerg KLEFFMANN, Laufs S.)
- University of Alberta, Canada (John WILSON)

**National Partners**
- ARVALIS (Xavier Foueillassar, …)
- INRA Ephyse (Eric LAMAUD, Sylvain DUPONT, …)
- INRA Eco-Inov (Frédérique ANGEVIN, …)
- INRA Rennes (Chris FLECHARD, …)
- INRA Lusignan (Abad CHABBI & co)
- Université Bordeaux (Stéphane GLOCKNER, …)

**Scientists**
- Carole BEDOS (CR)
- Pierre CELLIER (DR)
- Sophie GENERMONT (CR)
- Laurent HUBER (DR)
- Patricia LAVILLE (IR)
- Erwan PERSONNE (MC)
- Sébastien SAINT-JEAN (MC)
- Andrée TUZET (CR)

**Ingeniors and technicians**
- Hervé AUTRET (TR)
- Céline DECUQ (AI)
- Brigitte DURAND (TR)
- Olivier FANUCCI (TR)
- Dominique FLURA (AI)
- Jean-Christophe GUEUDET (TR)
- Sylvie MASSON (TR)
- Nicolas MASCHER (IE)
- Olivier ZURFLUH (IE)

**& all the LAB**

**Structurating Projects**
- GRAMNIAE (FP6)
- CARBOEUROPE (FP7)
- NITEOEUROPE (FP7)
- SAMBA (NERC)
- PHOTOONA (CNRS-DFG)
- ECLAIRE (FP7)
- INGOS (FP7)
- POLLEN (INSU-CNRS)
- BIOPOLLATM (ADEME, Ministère, …)
- R2DS (DIM Ile de France)

**Et**
- Merci à
- Sabine,
- Charlotte,
- Anais,
- Valentin
- Elisabeth
- & Michel