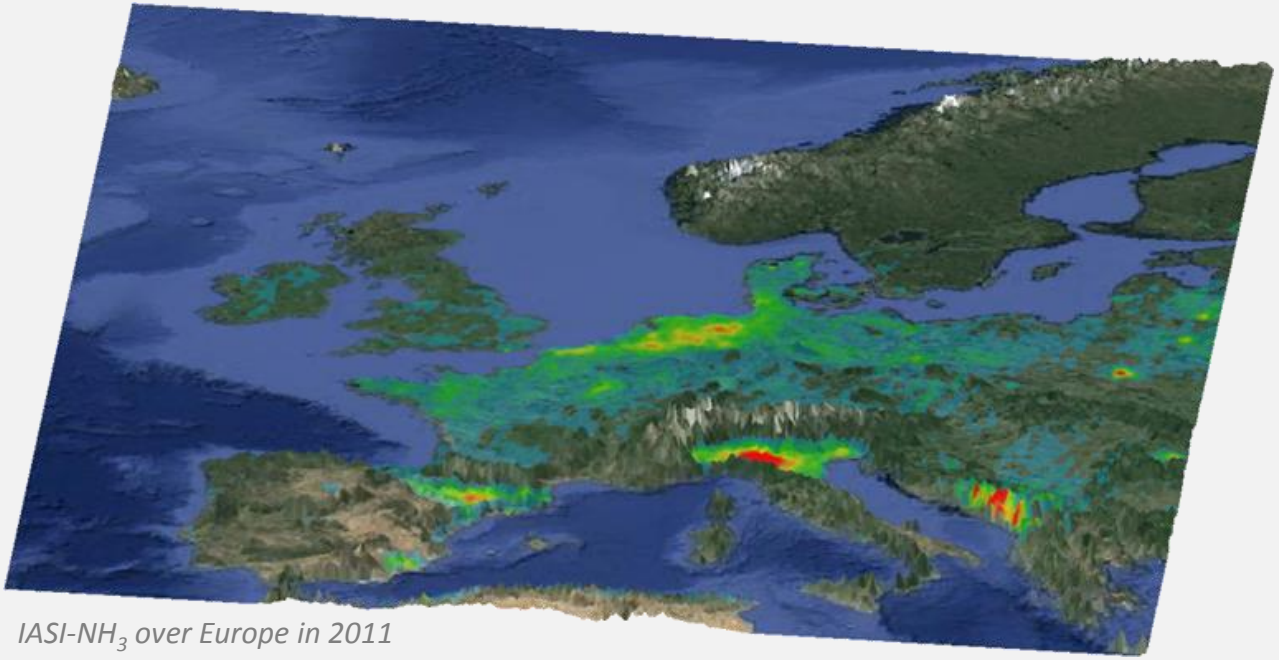
Worldwide spatial and temporal ammonia (NH_3) variability revealed by IASI



IASI-NH_3 over Europe in 2011

M. Van Damme
L. Clarisse, S. Whitburn, C. Clerbaux, P.-F. Coheur

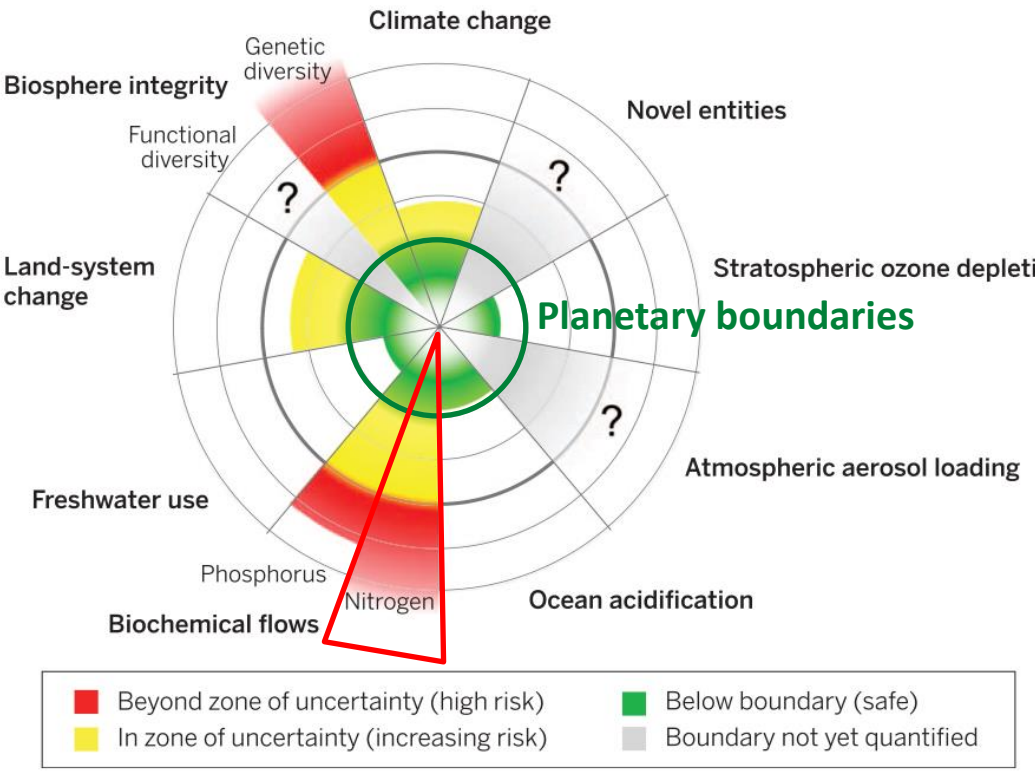
ULB LATMOS

N cycle perturbations

« Planetary boundaries »

="safe operating space for global societal development with respect to the Earth system and are associated with the planet's biophysical subsystems or processes"

N cycle is one of the main Earth-system processes for which the control variable has been exceeded



Earth-system process	Control variable	Planetary boundary (zone of uncertainty)	Current value of control variable
Biogeochemical flow: N cycle	Industrial and intentional biological fixation of N	**62** Tg N yr^{-1} (62–82 Tg N yr^{-1})	**~150** Tg N yr^{-1}

(Rockström *et al.*, Nature 2009; Steffen *et al.*, Science 2015)

N cycle perturbations

PM formation

La pollution à Paris aussi nocive que le tabagisme passif

LE MONDE | 24.11.2014 à 10h18 • Mis à jour le 25.11.2014 à 12h09 |

Par Sophie Landrin et Laetitia Van Eeckhout

© LeMonde.fr

China's largest algal bloom turns the Yellow Sea green

The algae, which can suffocate marine life, is thought to be caused by pollution from agriculture and industry

Karl Mathiesen

Thursday 4 July 2013 17.46 BST

© The Guardian

Eutrophication of costal water

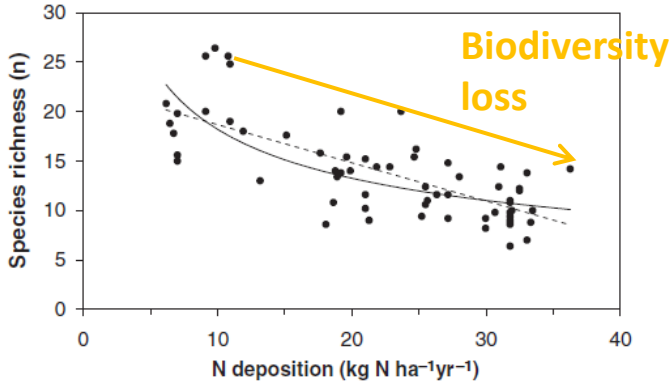
Les algues vertes gagnent les côtes de Normandie

Alerte : les algues vertes qui prolifèrent dans le surplus d'azote gagnent nos côtes. Dans le Calvados, à Grandcamp-Maisy, près de 1 000 tonnes d'algues ont déjà été relevées.

Mise à jour : 10/07/2014 à 12:33 par La Rédaction

© normandie-actu.fr

Biodiversity loss



(European Nitrogen Assessment, 2011)

Haber-Bosch process (1908): $N_2 + 3\,H_2 \rightarrow 2\,NH_3$

Inefficient use of Nr created:

80% of N consumed is lost!

> 2 x Nr introduction in the environment in comparison with before the industrial revolution

→ **Numerous environmental impacts**

WATER BODIES

AIR QUALITY

CLIMATE CHANGE

TERRESTRIAL ECOSYSTEMS

+

Nitrogen Cascade

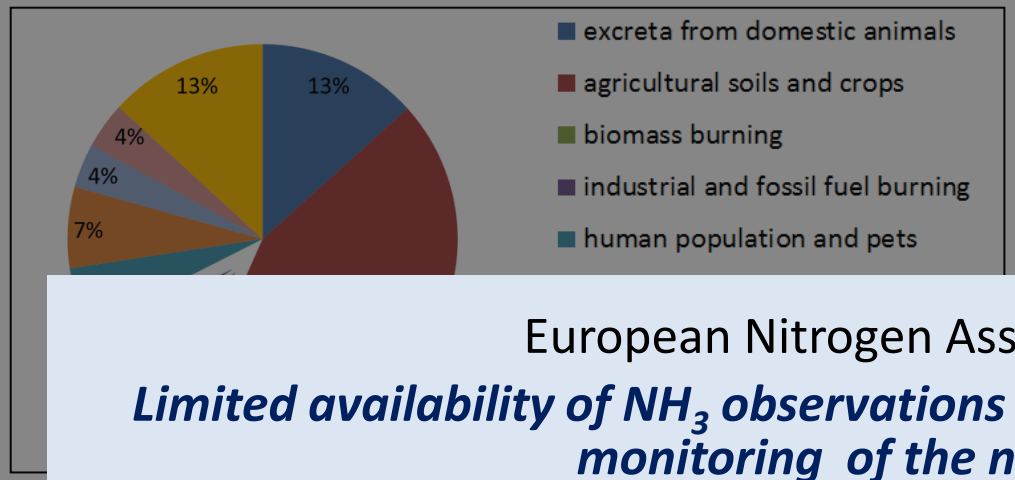
(Galloway et al., 2003)

N cycle perturbations

NH_3 in the atmosphere

65.4 Tg N released in 2008 (Sutton *et al.*, 2013) → **dominates global emission of total reactive nitrogen**

Sources:



- Uncertainties **> ± 30%**
- **Large variations at national/regional scale**

Sinks:

- PM formation

European Nitrogen Assessment, 2011:

Limited availability of NH_3 observations is currently a barrier for effective monitoring of the nitrogen cycle

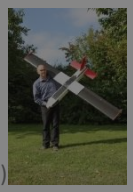
→ **GLOBAL NH_3 MEASUREMENTS FROM IASI**

1. Development of a retrieval algorithm for NH_3
2. Geophysical analyses of NH_3 distributions and time-series

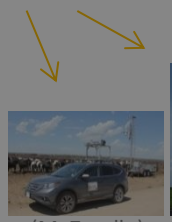
Me rk:

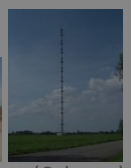
- f

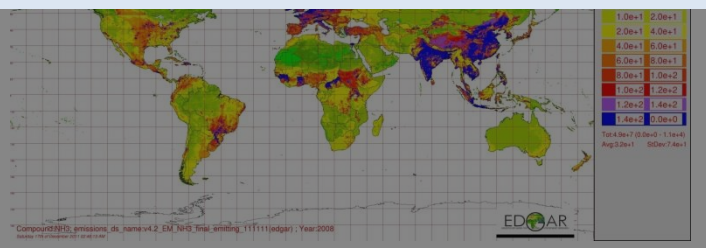
- F
- CIMS: Chemical Ionization Mass Spectrometer
- DOAS: Differential Optical Absorption Spectroscopy


(K. Skjøth)

(NOAA)


(M. Zondlo)


(Cabauw)

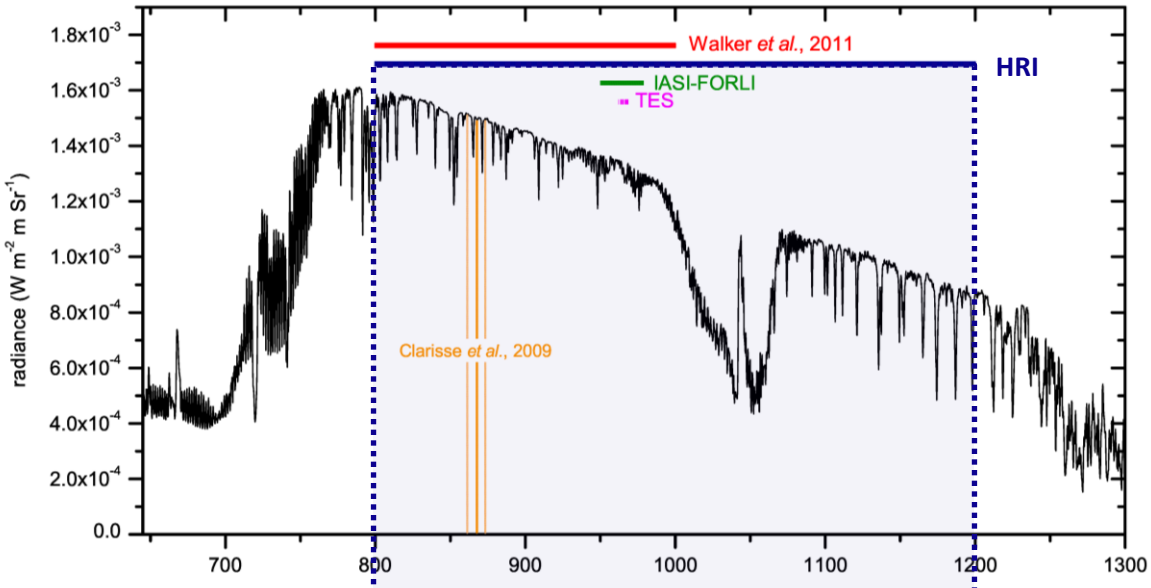


(EDGAR v4.2, 2011)

- Limited by the lack of reliable inventories/ observations

Development of a retrieval algorithm for NH_3

Method (1) Calculation of the Hyperspectral Range Index (HRI)

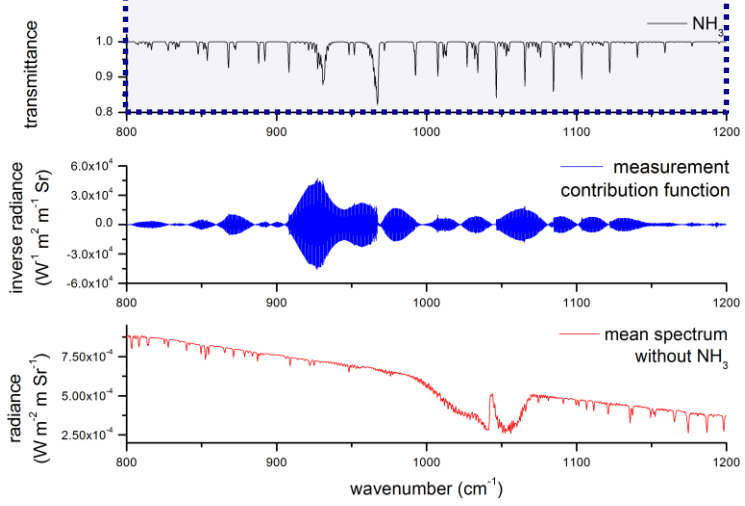


quantity representative of the NH_3 abundance

y measured spectrum

$$HRI = G\,(y - \bar{y})$$

NH_3 ν_2 vibrational band



(Van Damme *et al.*, ACP 2014)

Jacobian

$$G = (K^T S_y^{obs} K)^{-1} K^T S_y^{obs\,-1}$$

measurement contribution function

variance/covariance matrix of the spectral ensemble

$$S_y^{obs} = \frac{1}{N-1} \sum_{j=1}^{N} (y_j - \bar{y})(y_j - \bar{y})^T$$

average of the spectral ensemble (N y_j IASI spectra without NH_3)

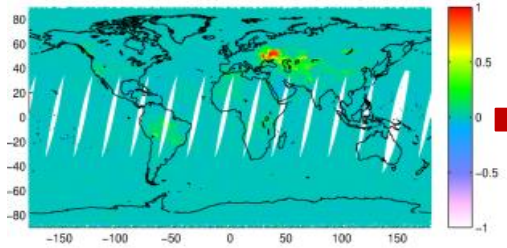
Development of a retrieval algorithm for NH_3

Method (2) From HRI to NH_3 column

15/08/2010

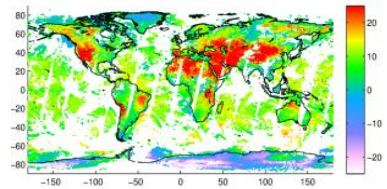
IASI L1C

HRI

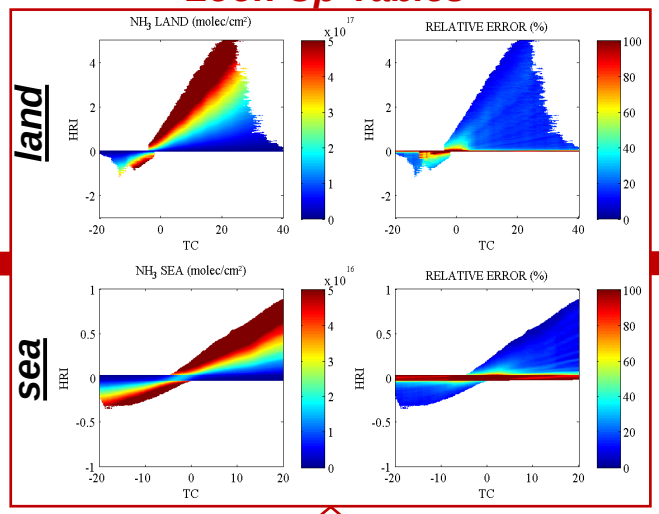


IASI L2

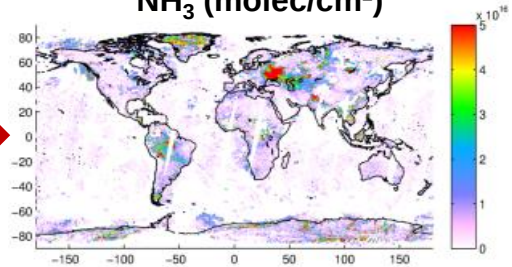
THERMAL CONTRAST (K)



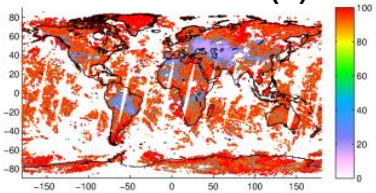
Look-Up-Tables



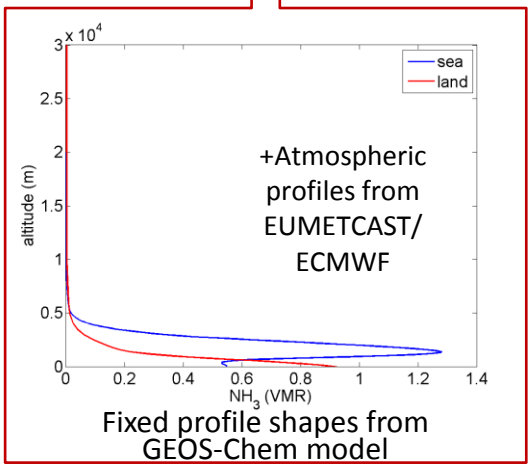
NH_3 (molec/cm²)



RELATIVE ERROR (%)



Line-by-line RTM



Fixed profile shapes from GEOS-Chem model

(Van Damme *et al.*, ACP 2014)

+

- Near-real time processing
- Low computational resources
- Increased sensitivity
- Error characterization

-

- Only clear-sky scene processed
- Uncertainties depend on the amount of NH_3 and the thermal contrast
- Fixed profile shapes

Development of a retrieval algorithm for NH_3

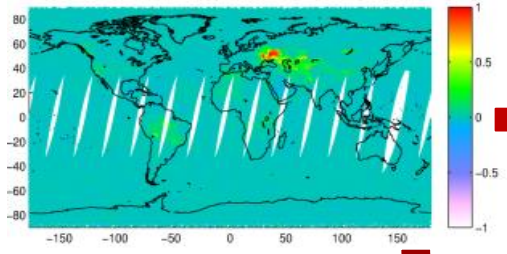
Method (2) From HRI to NH_3 column

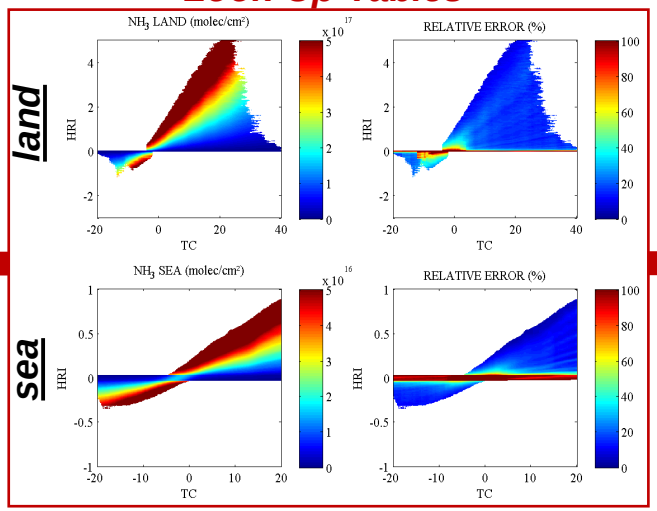
Look-Up-Tables

15/08/2010

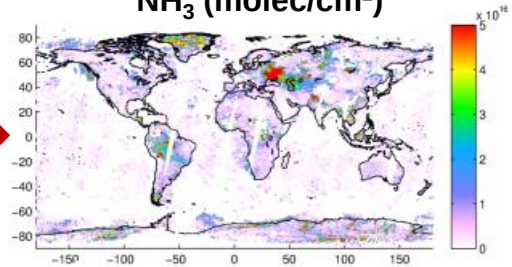
IASI L1C

HRI





NH_3 (molec/cm²)



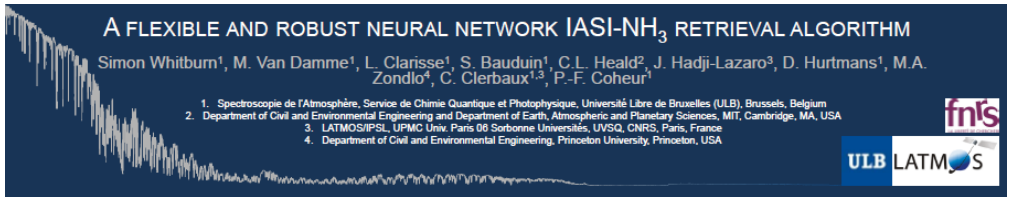
Artificial Neural Network!

$$NH_3 \text{ column} = HRI \times f(T, T_{surf}, P, H_2O, \sigma, z_0, \varepsilon, \text{angle})$$

(Whitburn *et al.*, accepted in JGR-atm)

Advantages:

- ✓ **Full atmospheric state** is taken into account as there is no limit on the number of input parameters
- ✓ **Full uncertainty analysis** is achieved by perturbing the input parameters
- ✓ **Reduced bias**, NN approach allowing negative columns
- ✓ **Flexible NH_3 profiles**

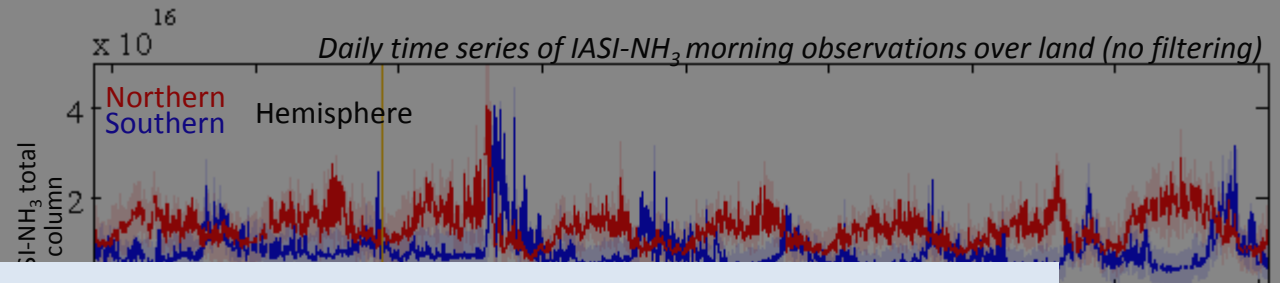


Poster S9-69

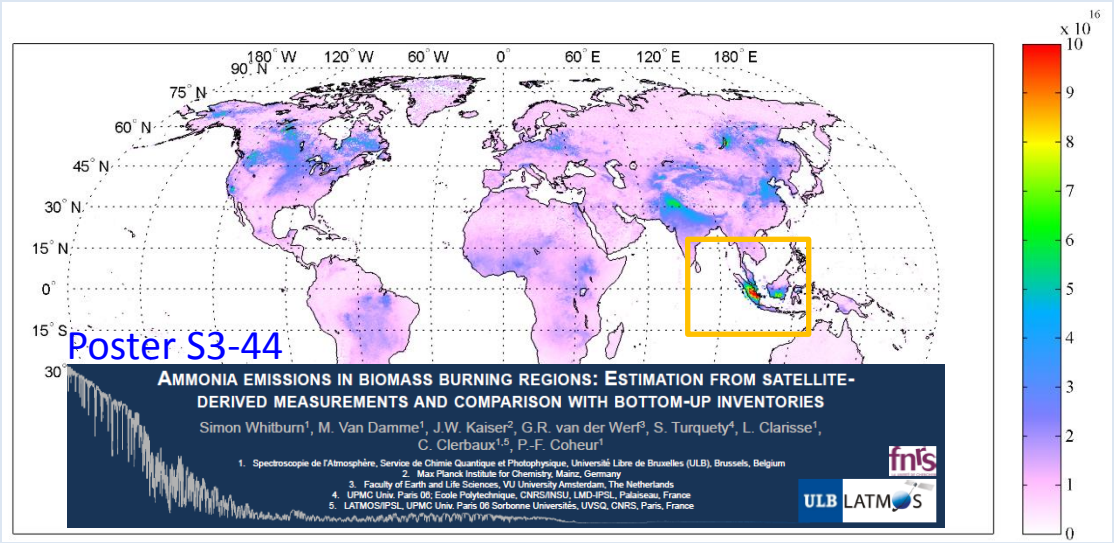
Development of a retrieval algorithm for NH_3

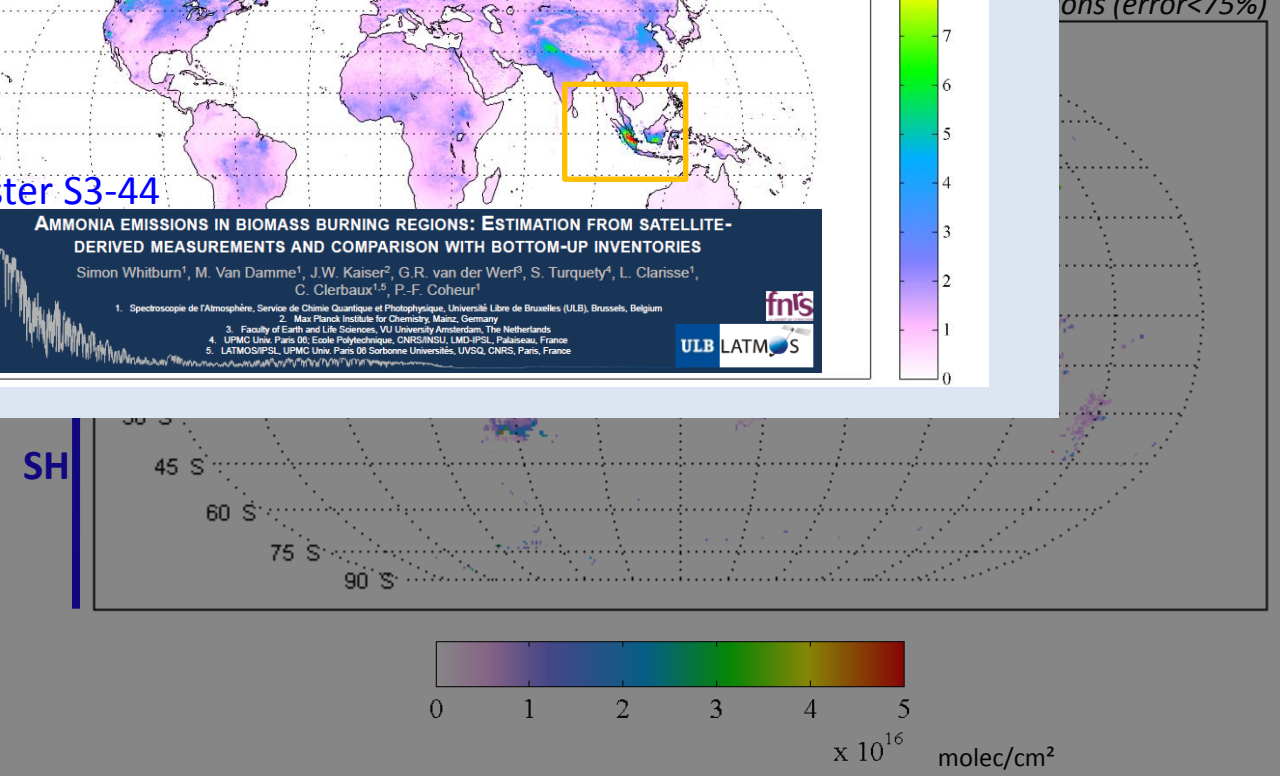
IASI-NH_3 data set

- Nov 2007
  → today
  (→ 2
- Higher columns
  Northern
  Hemisphere
- 2010 & 2015
  exceptional yea
  →T°
  →Fires



2015



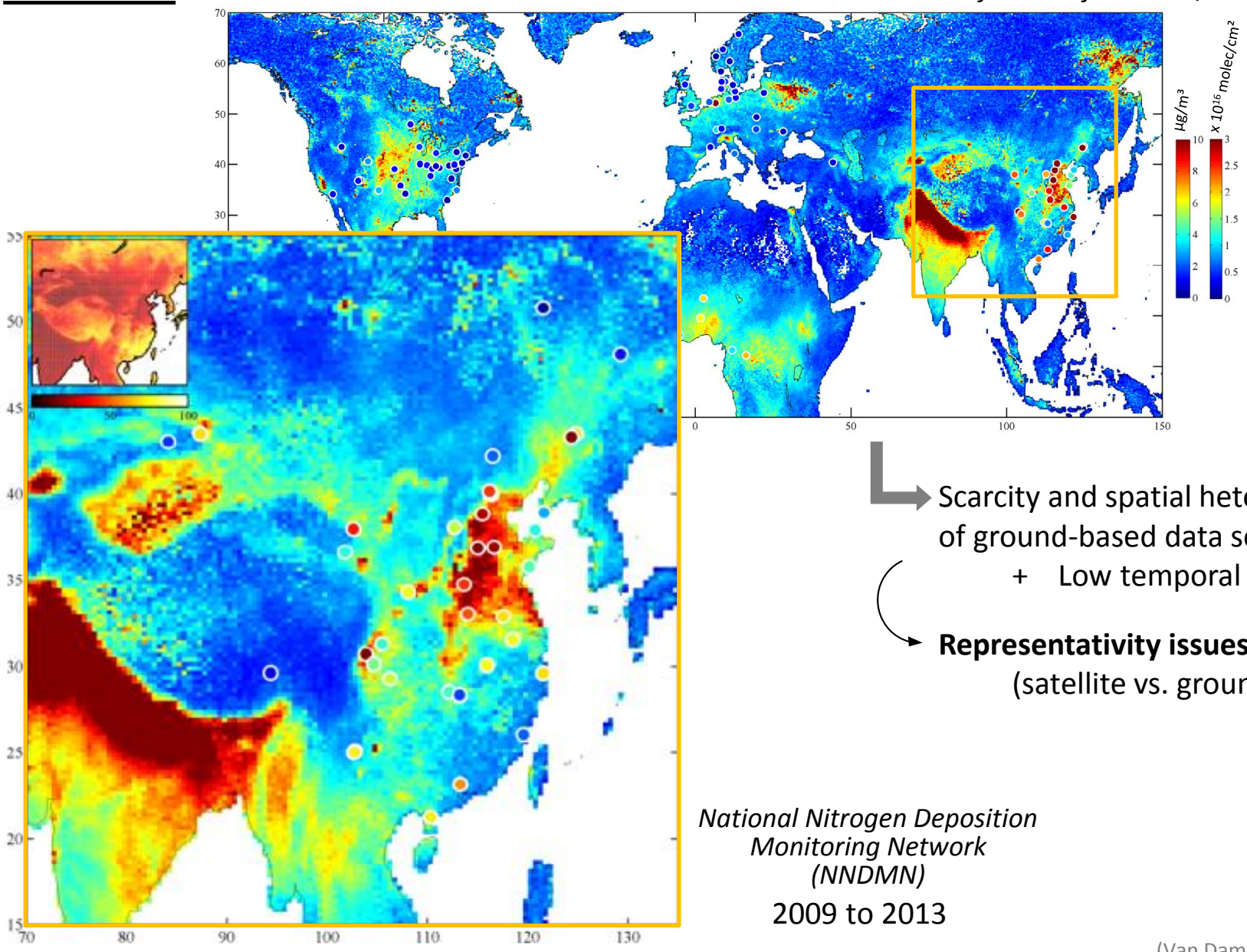


Development of a retrieval algorithm for NH_3

Validation

Ground-based observations

● = *observations for 2011 from IDAF, AMoN, EMEP and NNDMN networks*



→ Scarcity and spatial heterogeneity of ground-based data sets

\+ Low temporal resolution

→ **Representativity issues** (satellite vs. ground-based)

National Nitrogen Deposition Monitoring Network (NNDMN)

2009 to 2013

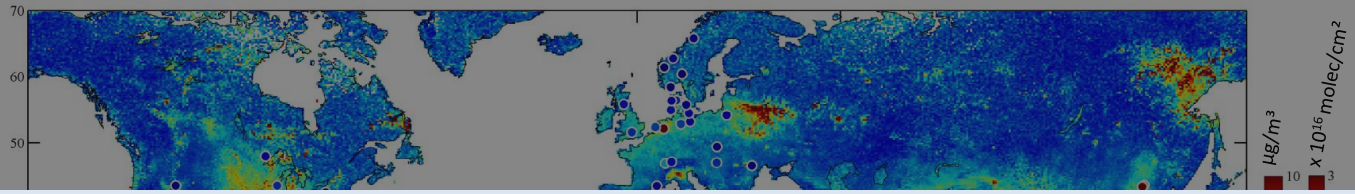
(Van Damme *et al.*, AMT 2015)

Development of a retrieval algorithm for NH_3

Validation

Ground-based observations

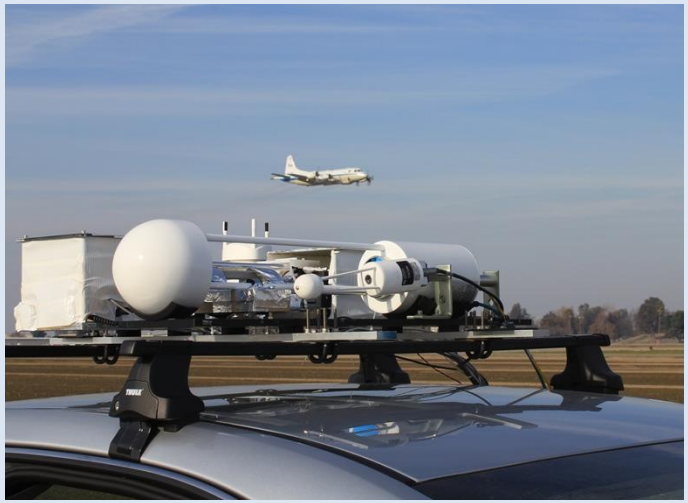
● = *observations for 2011 from IDAF, AMoN, EMEP and NNDMN networks*



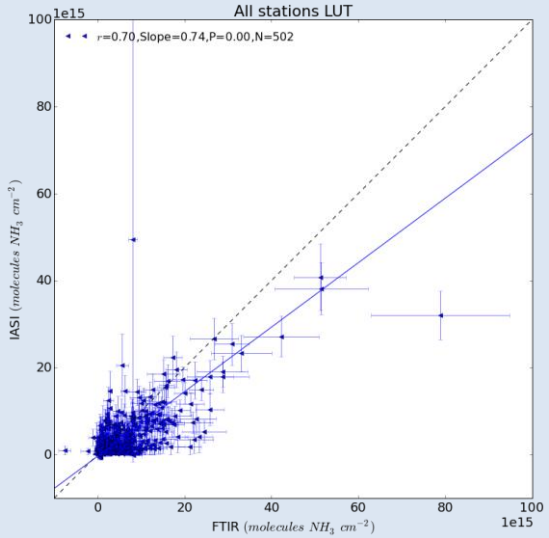
THURSDAY 14 APRIL

Session 10: Validation

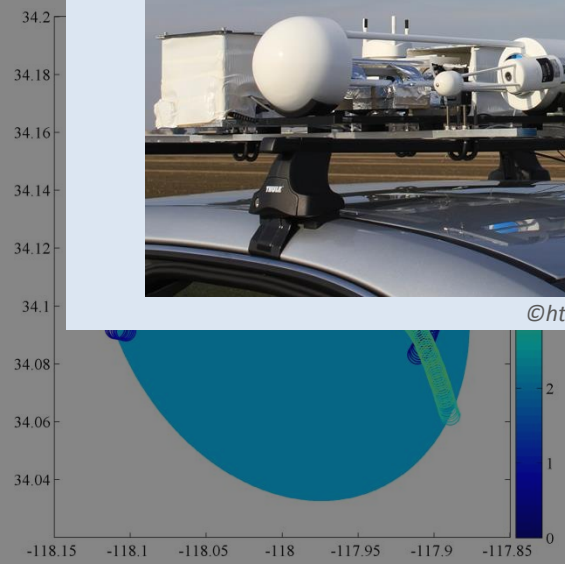
14:00 Validation of IASI NH3 columns at the single pixel scale from airborne- and ground-based measurements
Zondlo Mark (Princeton University)



©http://zondlo.princeton.edu



"An evaluation of IASI-NH_3 with ground-based FTIR measurements"
(Dammers *et al.*, ACPD 2016)



= 1515 NOAA WP-3D aircraft observations →2.65 ppbv

→ **Very good correlation obtained with airborne data set**

(Van Damme *et al.*, AMT 2015)

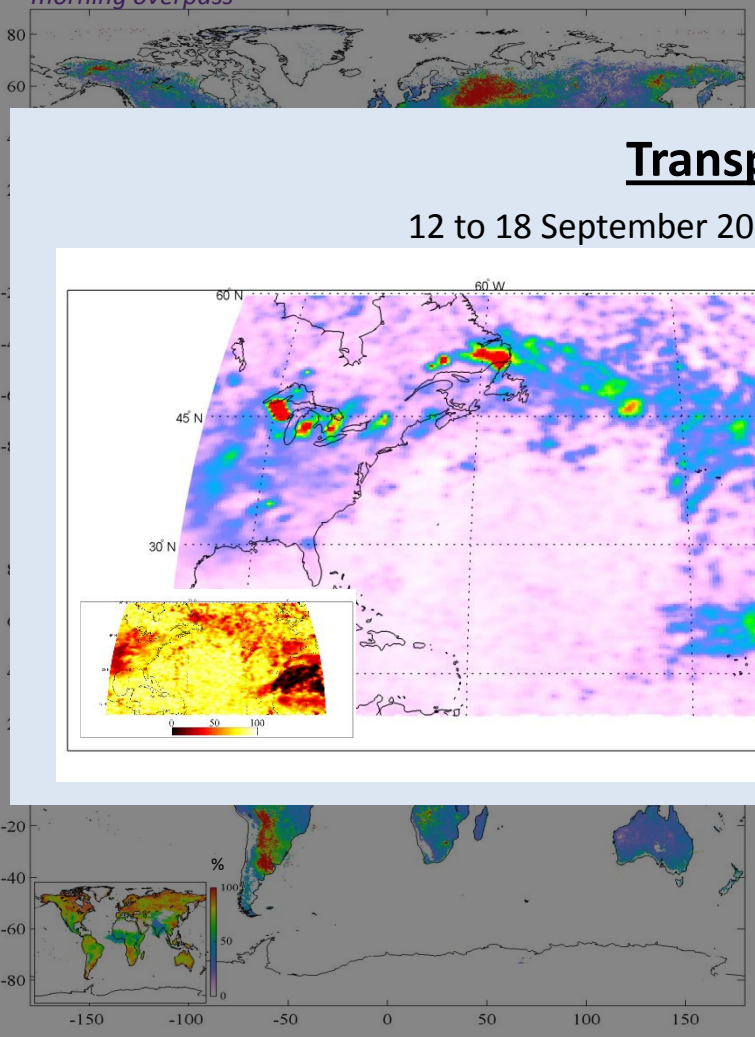
Geophysical analyses of NH_3 distributions & time-series

Global distributions

5-yr mean distributions (Nov-2007 → Oct-2012)
Mean columns associated with a relative error > 75%/58% (land/sea) have been filtered out

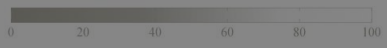
6-yr composite seasonal distributions over land (2008 → 2013)
Mean columns associated with a relative error > 75% have been filtered out

morning overpass



(Van Damme *et al.*, ACP 2014)

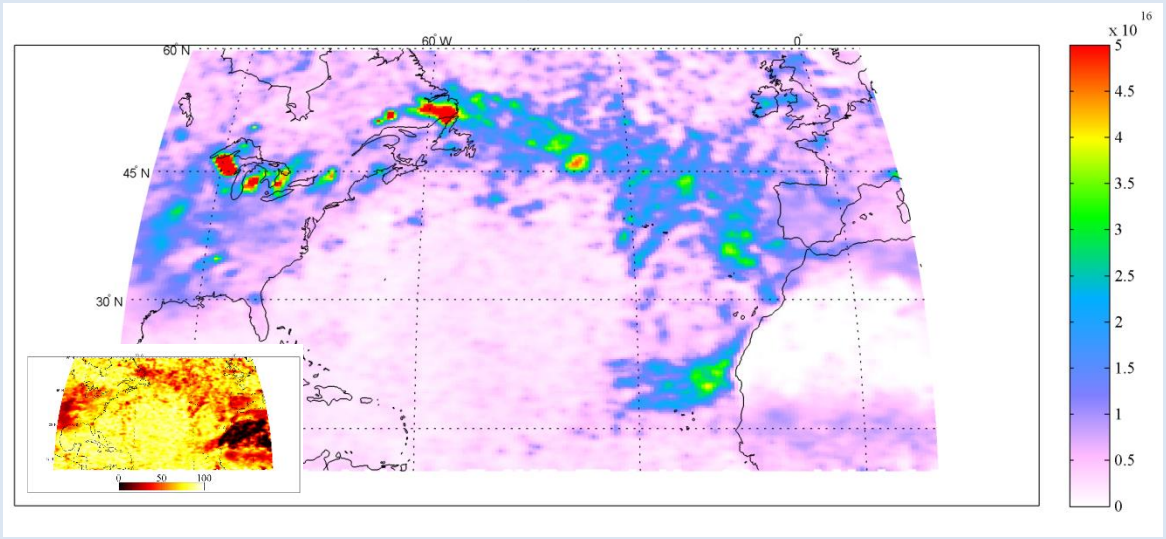
SON



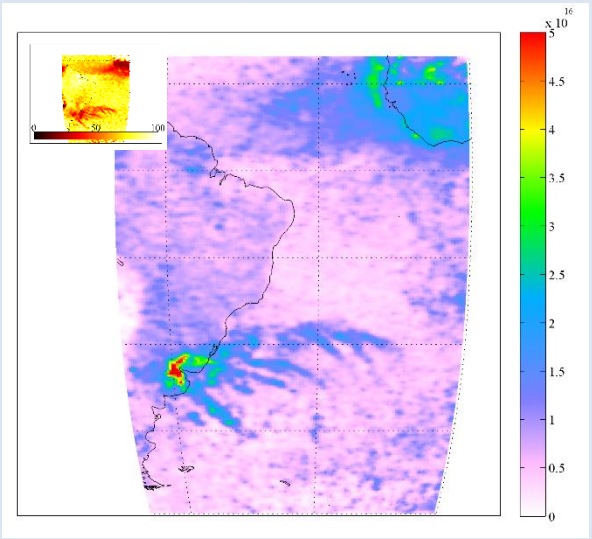
(Van Damme *et al.*, GRL 2015)

Transport over Atlantic Ocean

12 to 18 September 2011



April 2008



Geophysical analyses of NH_3 distributions & time-series

Global distributions

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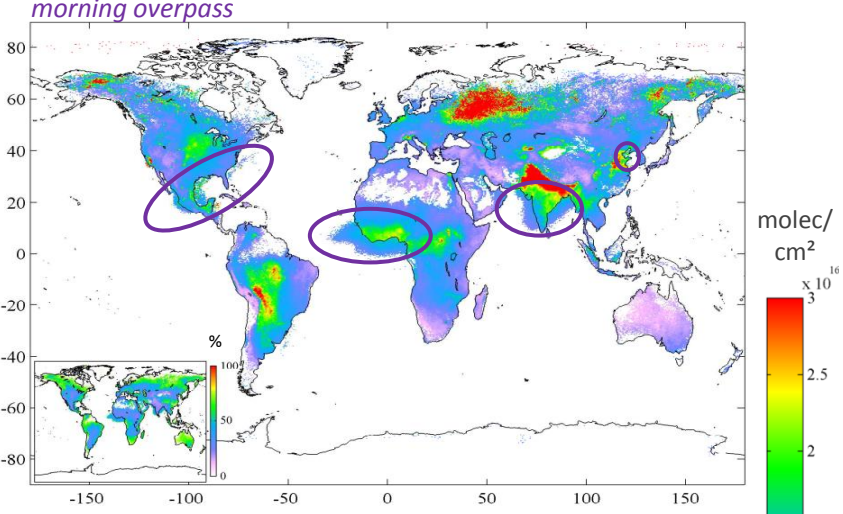
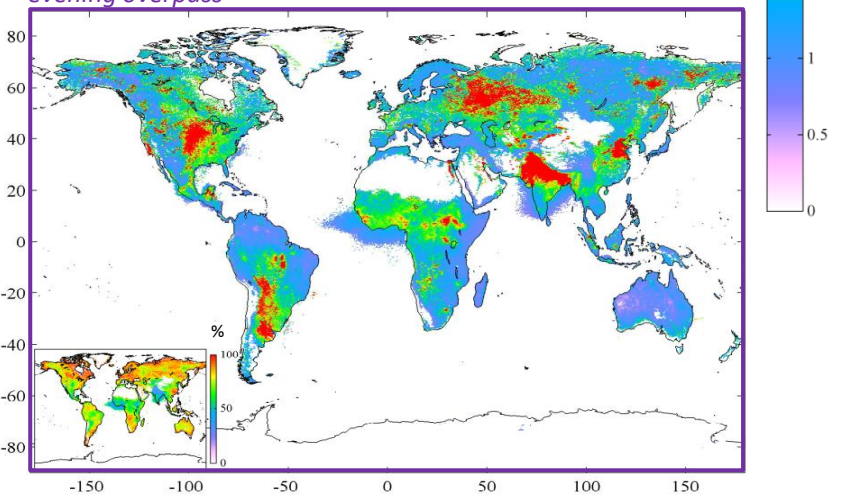
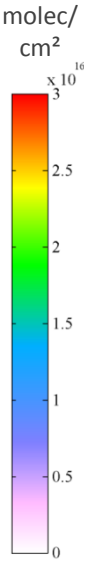


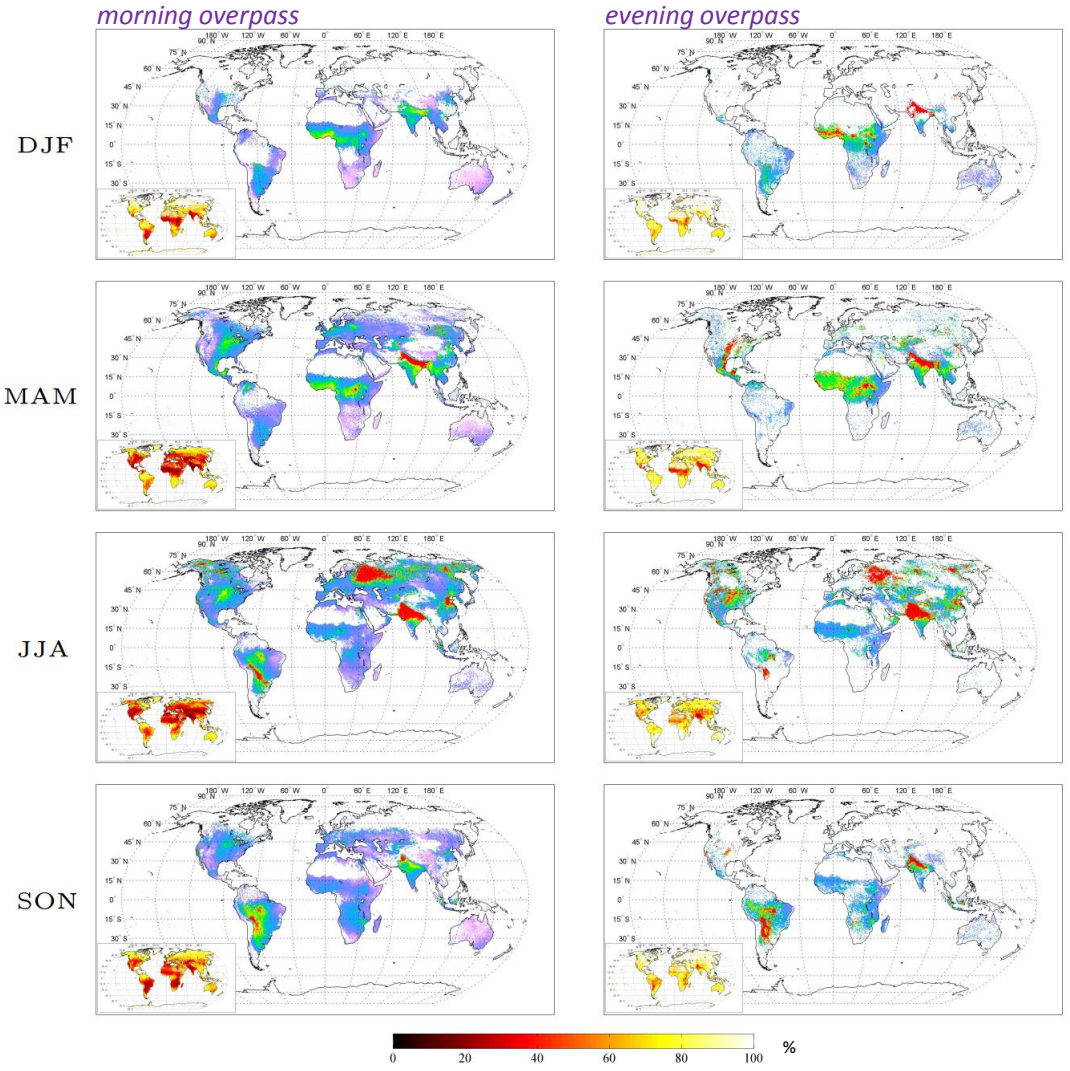
evening overpass





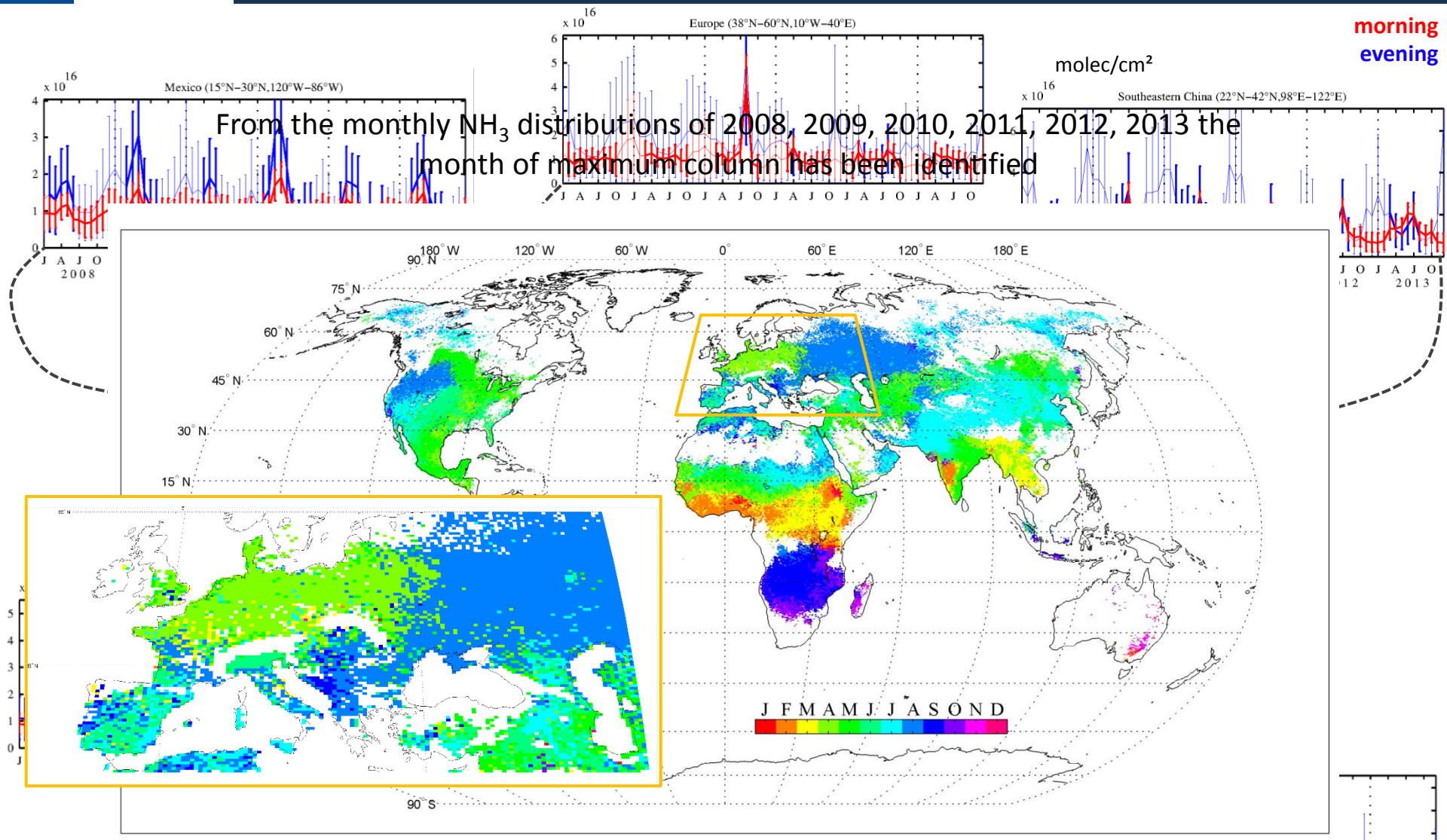
(Van Damme *et al.*, ACP 2014)

6-yr composite seasonal distributions over land (2008 → 2013)
Mean columns associated with a relative error > 75% have been filtered out



(Van Damme *et al.*, GRL 2015)

Geophysical analyses of NH_3 distributions & time-series



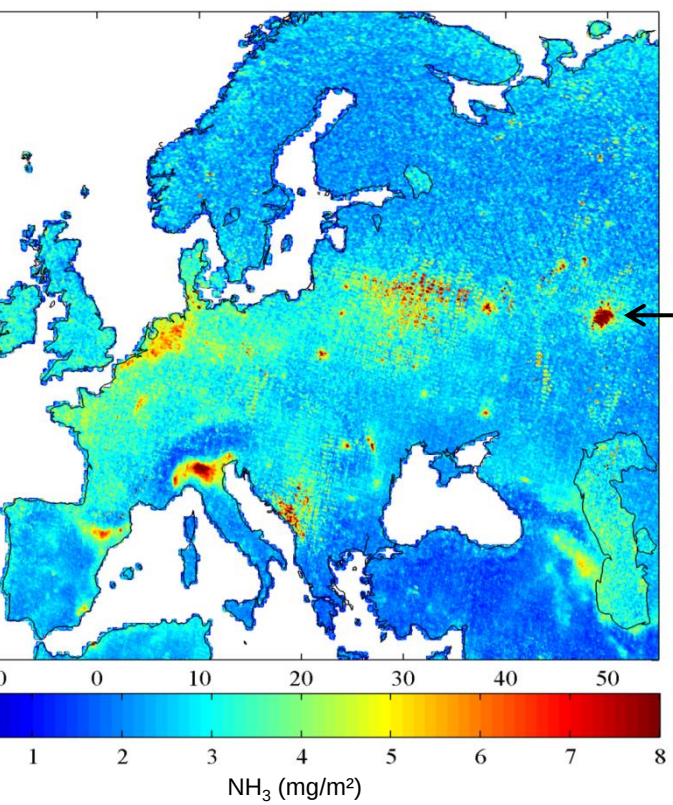
Geophysical analyses of NH_3 distributions & time-series

Model comparison

Inter-annual variability
4-yr comparison (2008→2011)

○ = industrial hot spots

LOTOS-EUROS (IASI) simulations | IASI observations | IASI error | IASI - LOTOS-EUROS



2008→2012 AM & PM weighted average
(excluding RU 2010 fire event period)

World's largest NH_3 producer in Togliatti, Samara Oblast, Russia

- 3 500*1000 metric NH_3 tons.yr^{-1}
- 85% exported
- World's longest NH_3 pipeline to Odessa, Black-sea

➢ **First time that industrial NH_3 sources are observed so clearly suggesting strong underestimation of emission in the inventories**

1.92
1.86
1.11
1.44

↗ L2 quality

(Van Damme *et al.*, JGR-Atm 2014)

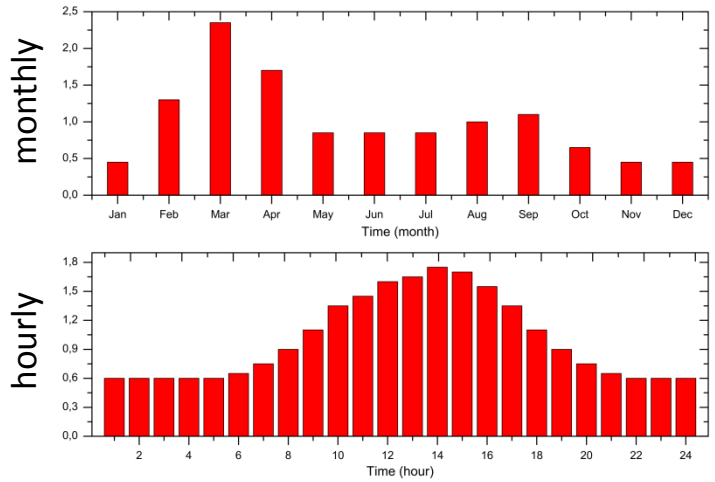
Geophysical analyses of NH_3 distributions & time-series

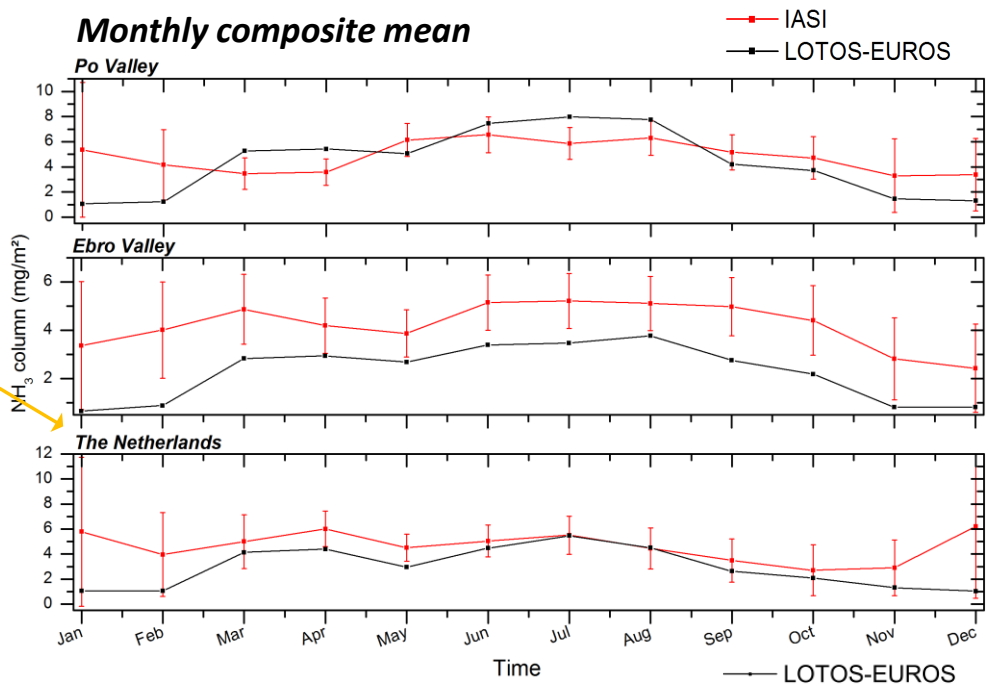
Model comparison

Intra-annual variability
4-yr comparison (2008→2011)

(Van Damme *et al.*, JGR-Atm 2014)

Fixed temporal profile for agricultural emissions used in the model

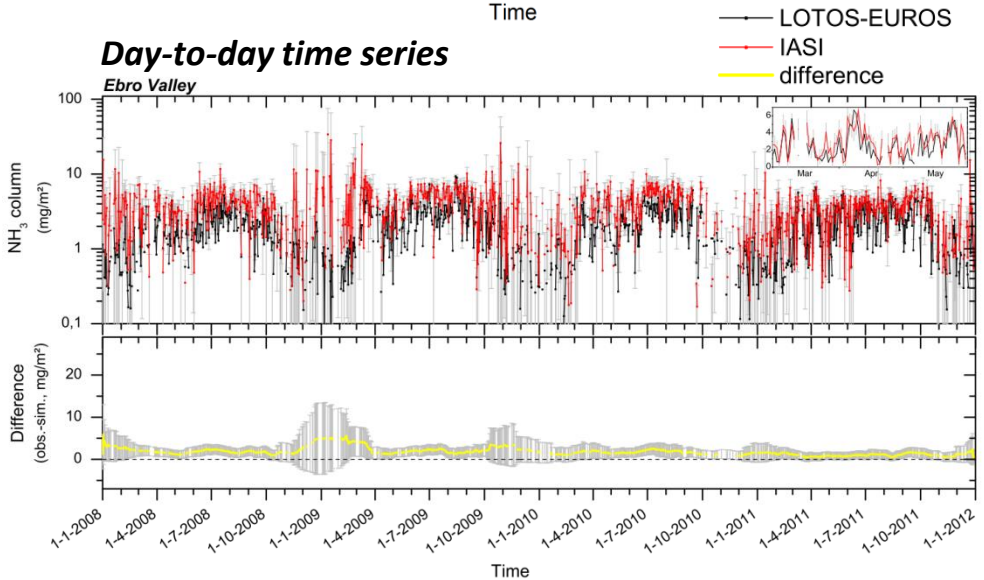




Emission timing and gridding assessment:

- Po Valley: recurring feature likely linked with wrong timing
- Ebro Valley: underestimated emission and/or misrepresentation of the diurnal profile
- Netherlands: best agreement (magnitude & timing)

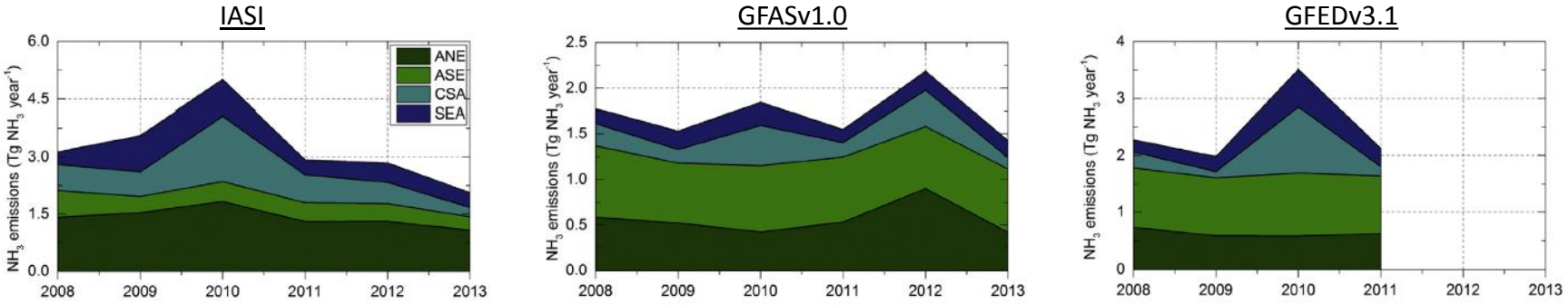
→expected as the fixed timing of the emissions are based on experimental data sets from this country



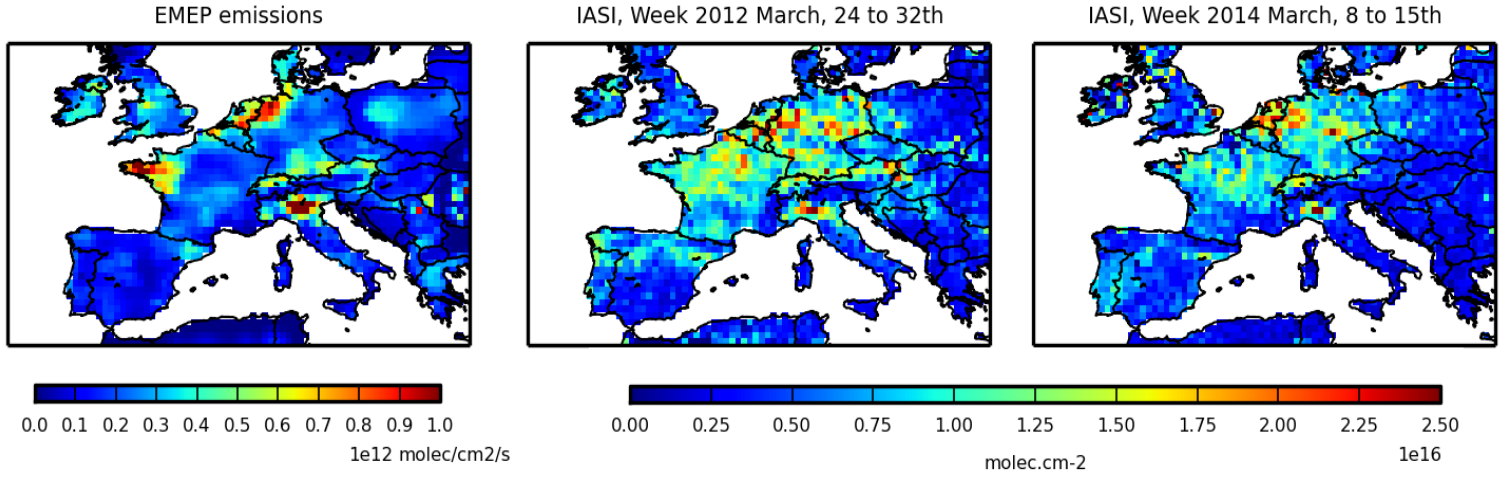
Geophysical analyses of NH_3 distributions & time-series

Applications

- Assessment of fire emissions inventories (Whitburn *et al.*, Atm. Env. 2015) +Poster S3-34



- Assimilation into CHIMERE model (Fortem-Cheney *et al.*, in revision for GRL) +Poster S3-33



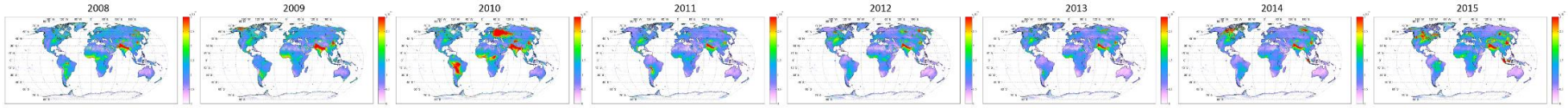
→ Significant impacts on (and increases of) PM concentrations

→ Improve the comparison with independent surface measurements of NH_3 and PM

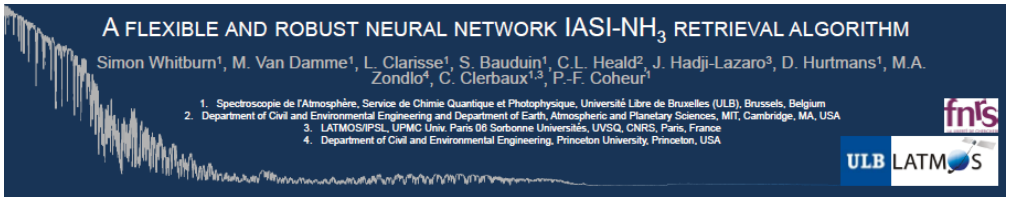
Final points

- **>8 years of global (bi-)daily IASI-NH_3 data (IASI-A and IASI-B) available in open access via Ether website**

Poster S4-50



- Flexible and robust **neural network** approach has been applied on IASI spectra



Poster S9-69

- Validation of IASI-NH_3: **Talk at 14:00 on Thursday (M. Zondlo)** Session 10

- Fire emissions assessment

Poster S3-44

- Assimilation into model

Poster S3-33

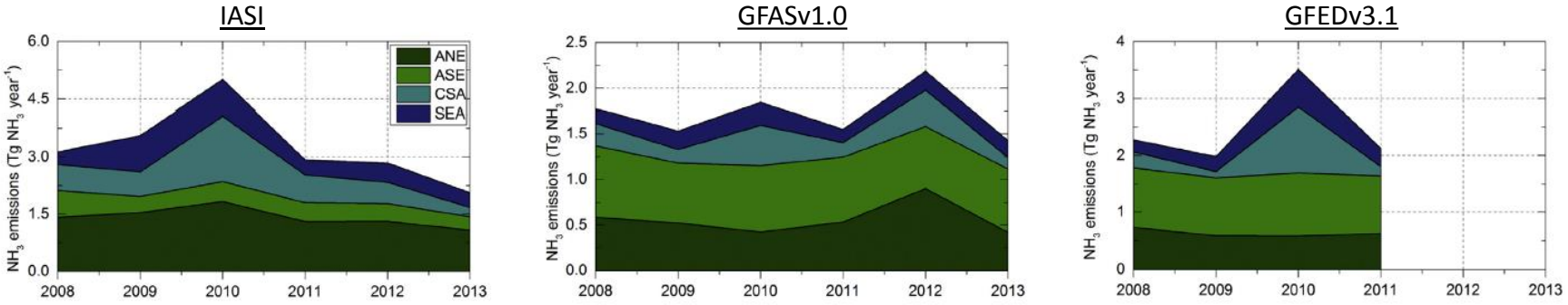
Thank you for your attention!

cnes CENTRE NATIONAL D'ÉTUDES SPATIALES · EUMETSAT · VU VRIJE UNIVERSITEIT AMSTERDAM · fnrs LA LIBERTÉ DE CHERCHER · éclaire Effects of climate change on air pollution impacts and response strategies for European ecosystems · TNO · belspo

Geophysical analyses of NH_3 distributions & time-series

Applications

- Assessment of fire emissions inventories (Whitburn *et al.*, Atm. Env. 2015) +Poster S3-34



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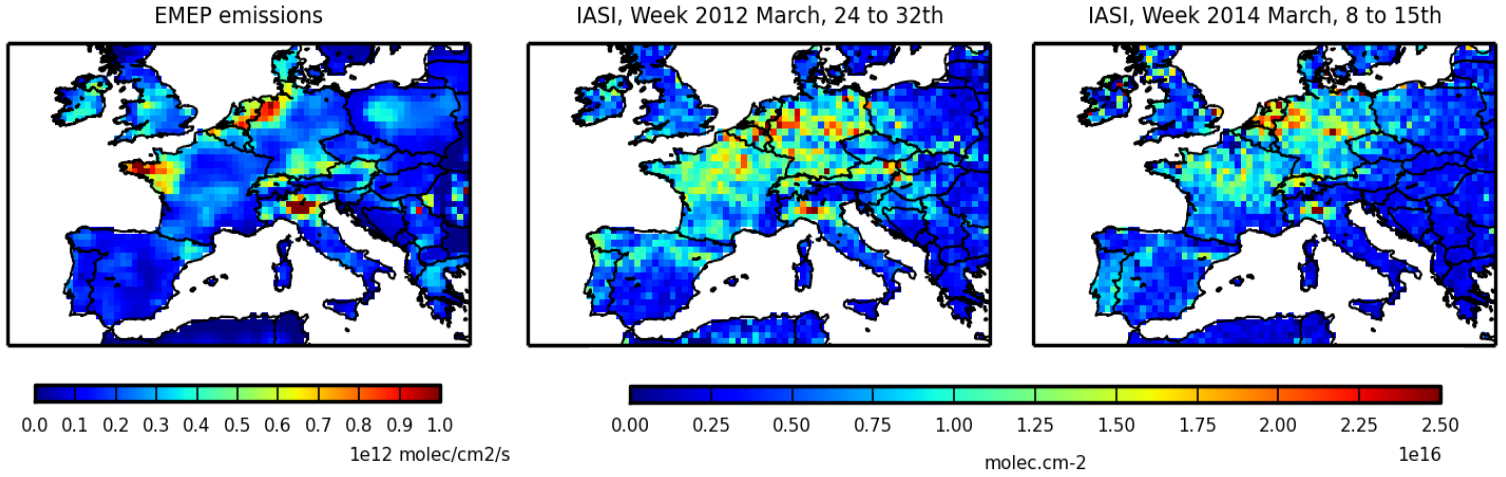
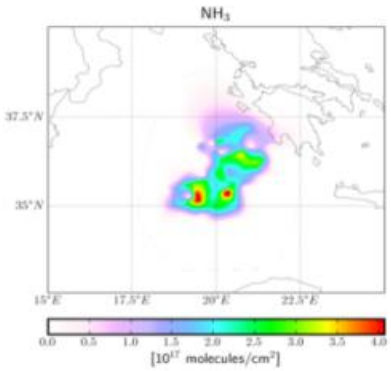


→ Significant impacts on (and increases of) PM concentrations

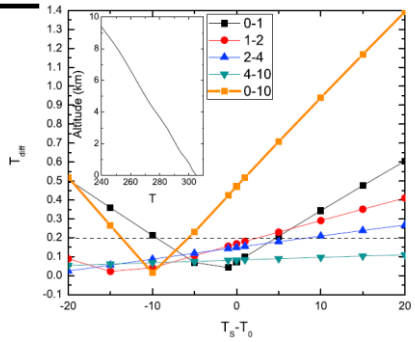
→ Improve the comparison with independent surface measurements of NH_3 and PM

Development of a retrieval algorithm for NH_3

Time-line of IASI-NH_3 retrievals



First detection
(Coheur *et al.*, ACP 2009)



Sensitivity study

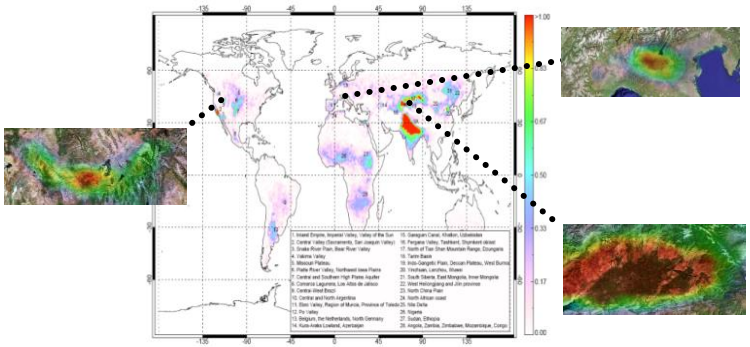
(Clarisse *et al.*, JGR 2010)

WE ARE HERE

HRI = Hyper-spectral Range Index

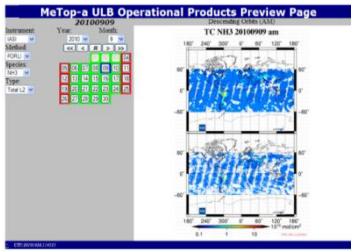
Hotspots

(Clarisse *et al.*, Nature Geo 2009)

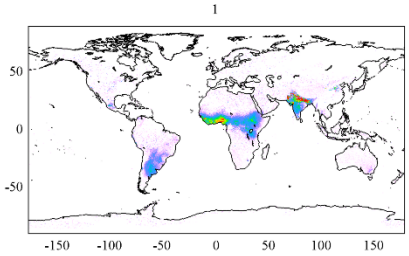


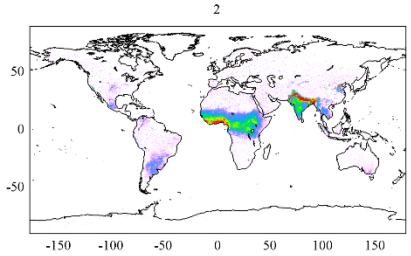
FORLI = Fast Optimal/ Operational Retrievals on Layers for IASI

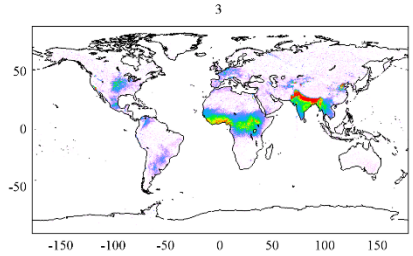
(Hurtmans et al., JQSRT 2012)

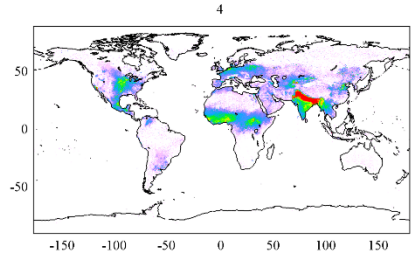


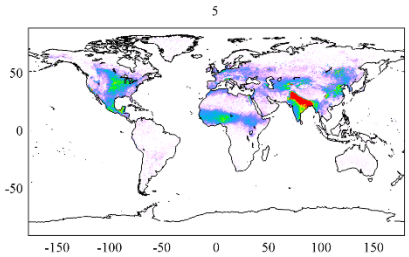
Geophysical analyses of NH_3 distributions & time-series

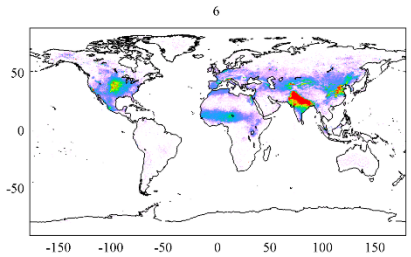


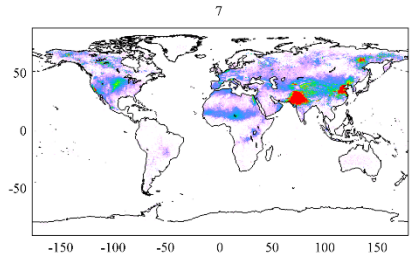


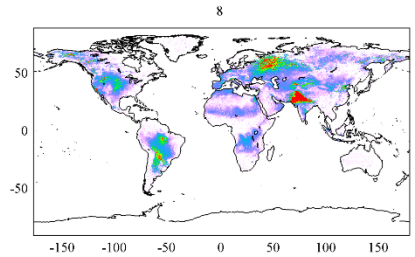


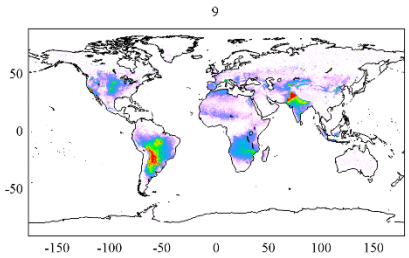


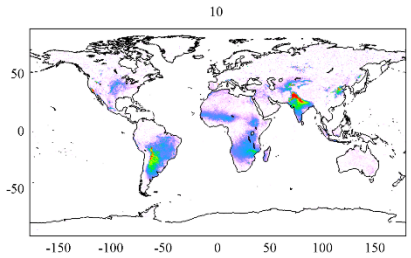


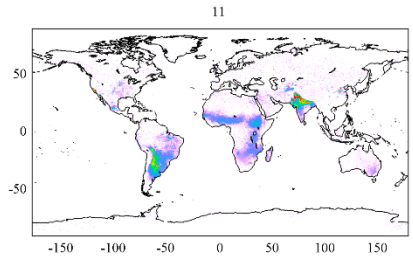


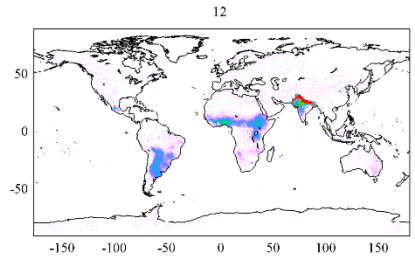




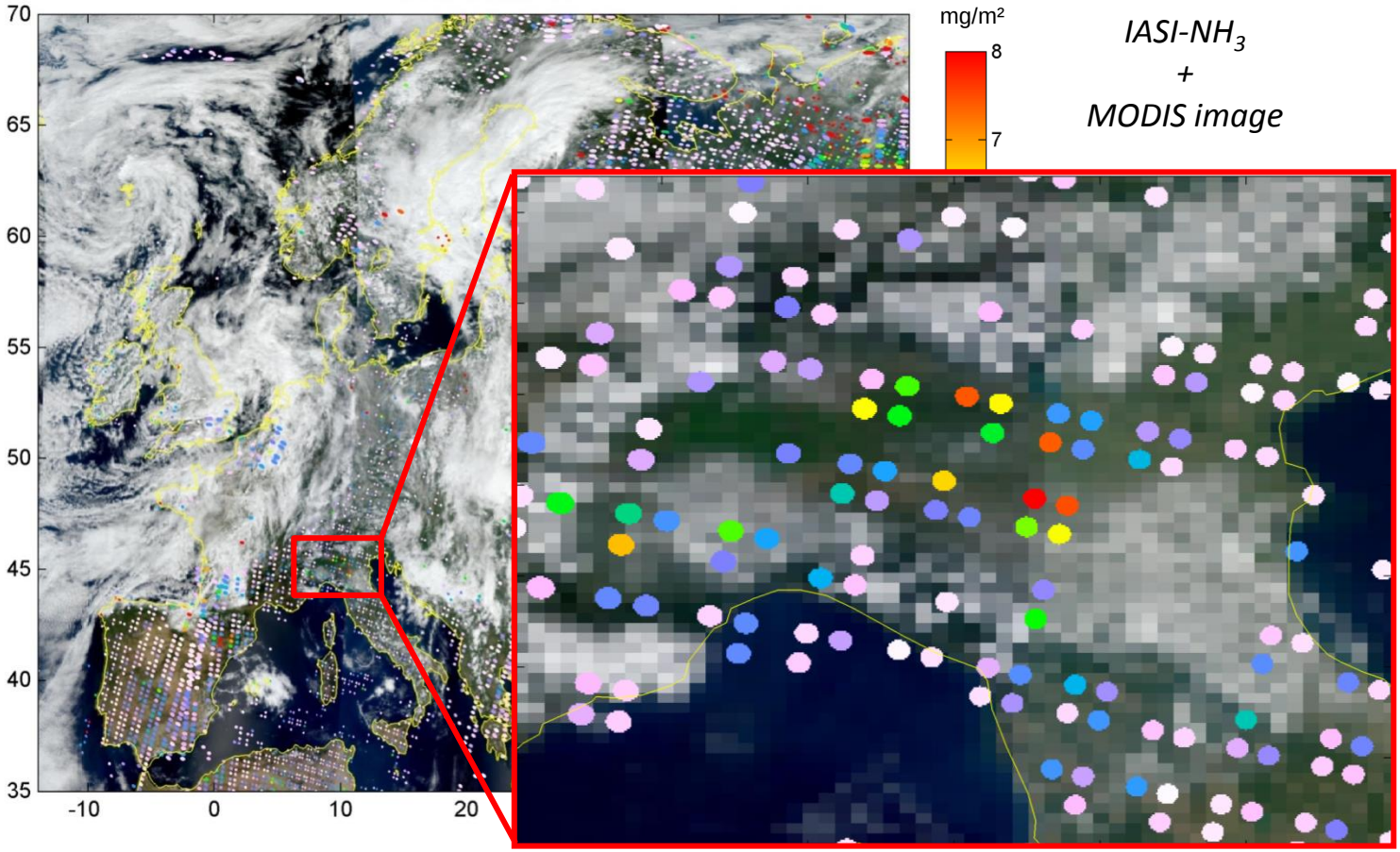








Geophysical analyses of NH_3 distributions & time-series



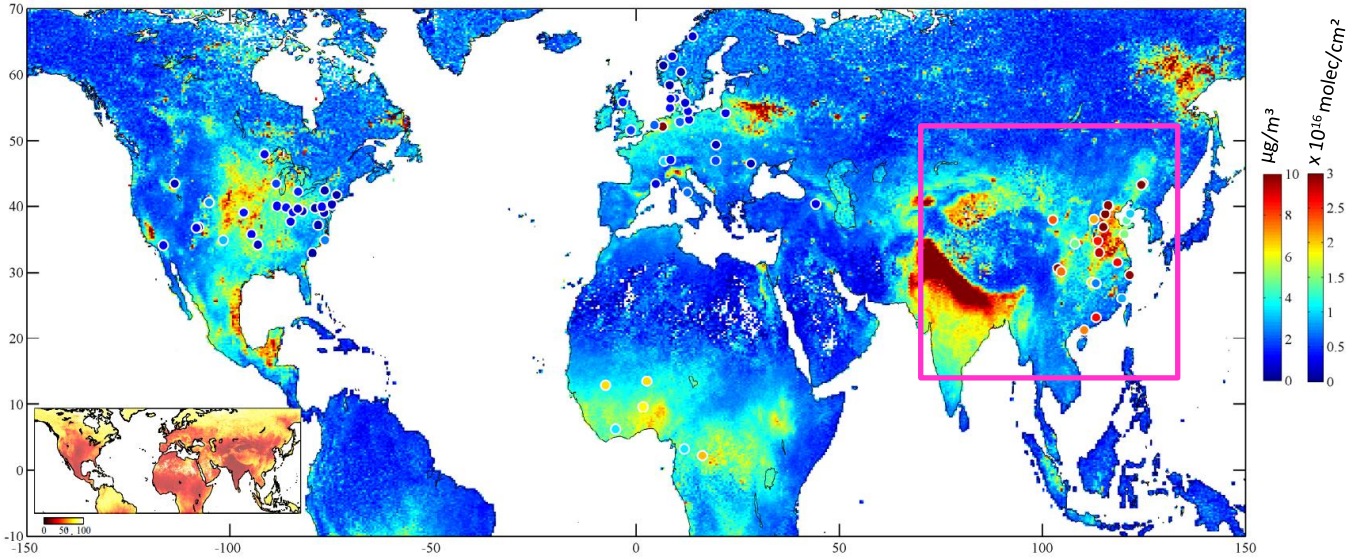
- ***cloudy data disregarded***
- ***large gap between footprints***

Development of a retrieval algorithm for NH_3

Validation

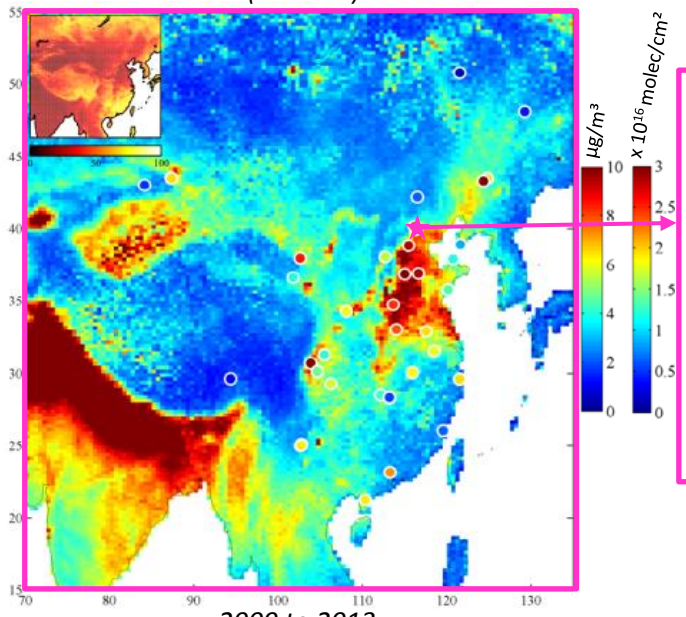
Ground-based observations

● = *observations for 2011 from IDAF, AMoN, EMEP and NNDMN networks*

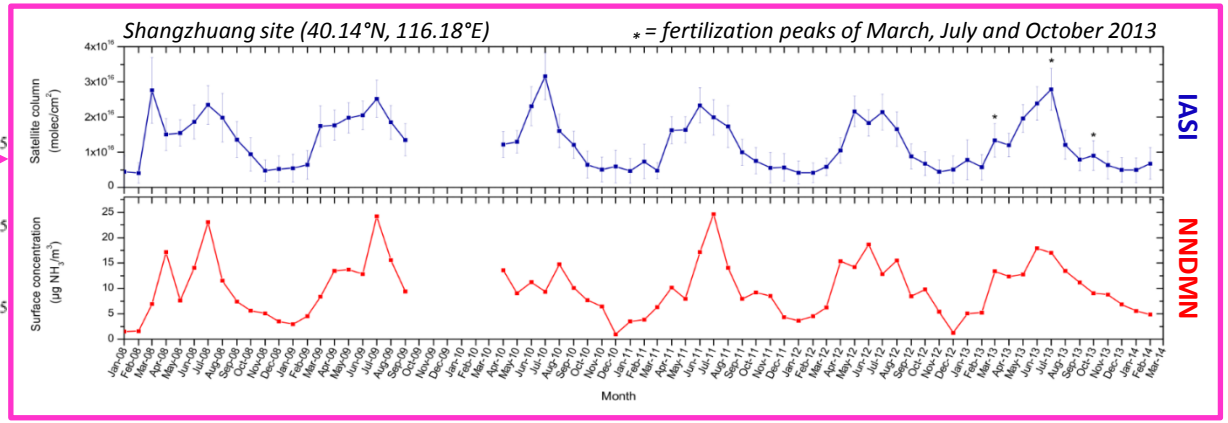


Qualitative comparison: IASI columns vs. surface abundances

National Nitrogen Deposition Monitoring Network (NNDMN)



2009 to 2013



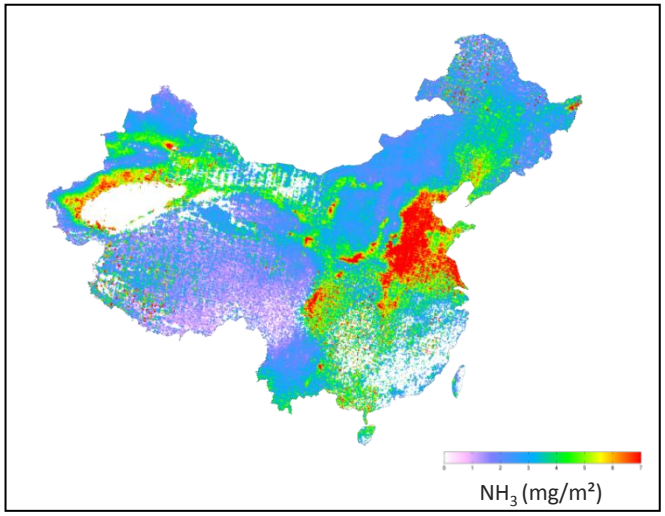
(Van Damme *et al.*, AMT 2015)

Geophysical analyses of NH_3 distributions & time-series

Satellite vs Emission Inventories

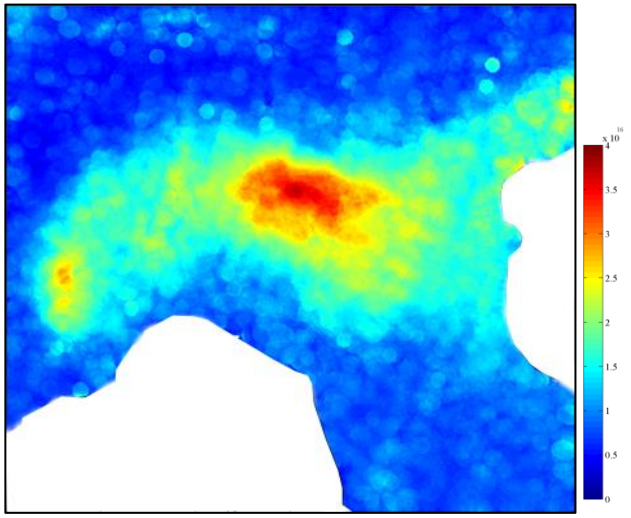
China

IASI total columns in 0.05ºx0.05º grid (mg/m²)



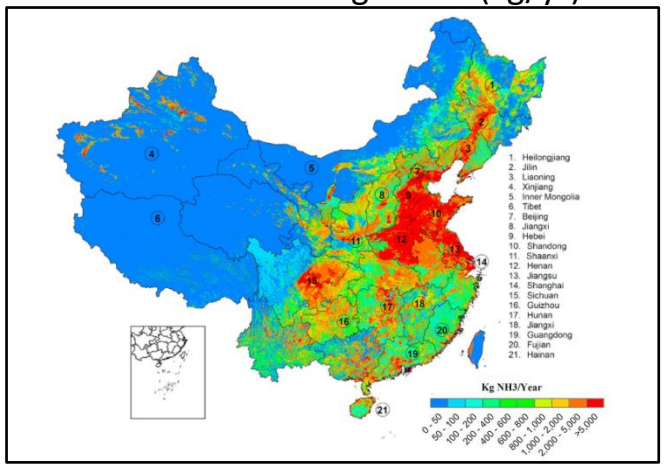
Po Valley

IASI total columns in 0.01ºx0.01º grid (molec/cm²)



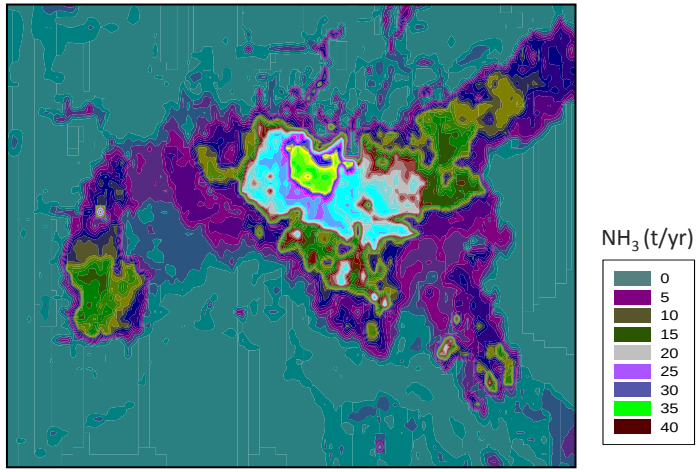
IASI satellite

Spatial distribution of ammonia emission in 1 km grid cell (kg/yr)



(Huang *et al.*, GBC 2012)

ARPA-ER inventory in 5 km grid cell (t/yr)

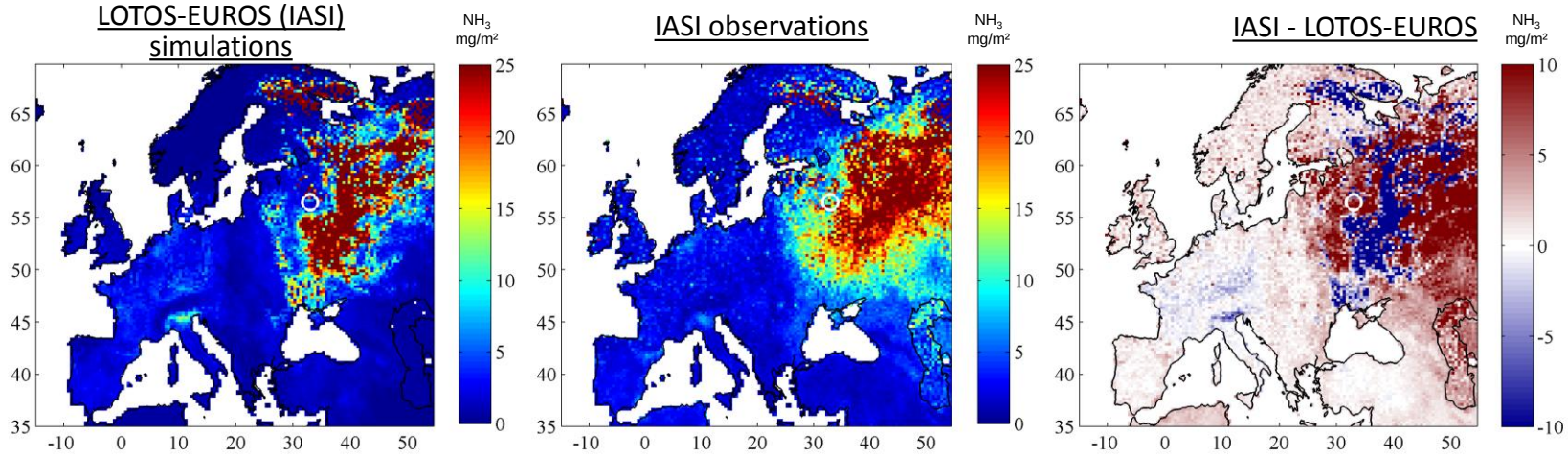


Emissions inventory

Geophysical analyses of NH_3 distributions & time-series

Russian fire episode in 2010

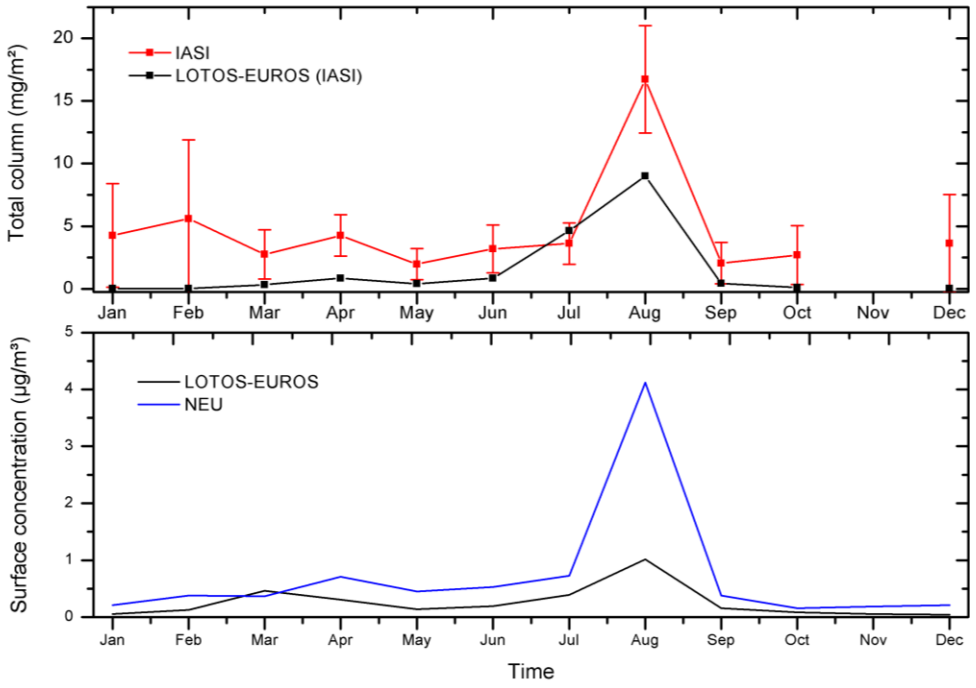
20 July to 20 August mean



Emission inventories

	NH_3 emitted from fires (Tg)	(% of total)
2008	0,096	1,51
2009	0,097	1,53
2010	0,98	13,54
2011	0,076	1,20

10x



- Transport not well accounted for in the model
- Missing emissions
- Height of injections (plume up to 12km)

  →Limitations from the fixed profile shape used for the IASI retrieval

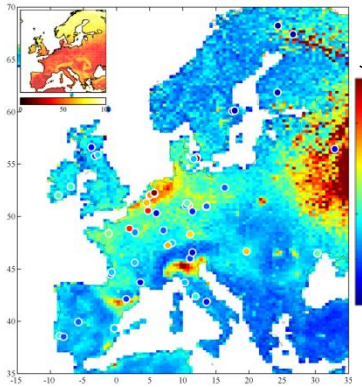
  →Limitations from the vertical extend (3.5 km) of LOTOS-EUROS model

Development of a retrieval algorithm for NH_3

Validation

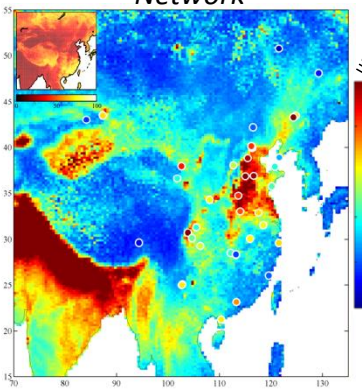
Ground-based observations

Nitro EUrope



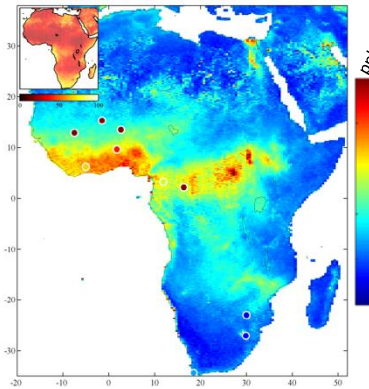
2008 to 2010

National Nitrogen Deposition Monitoring Network



2009 to 2013

IGAC/DEBITS/AFRICA



2008 to 2011

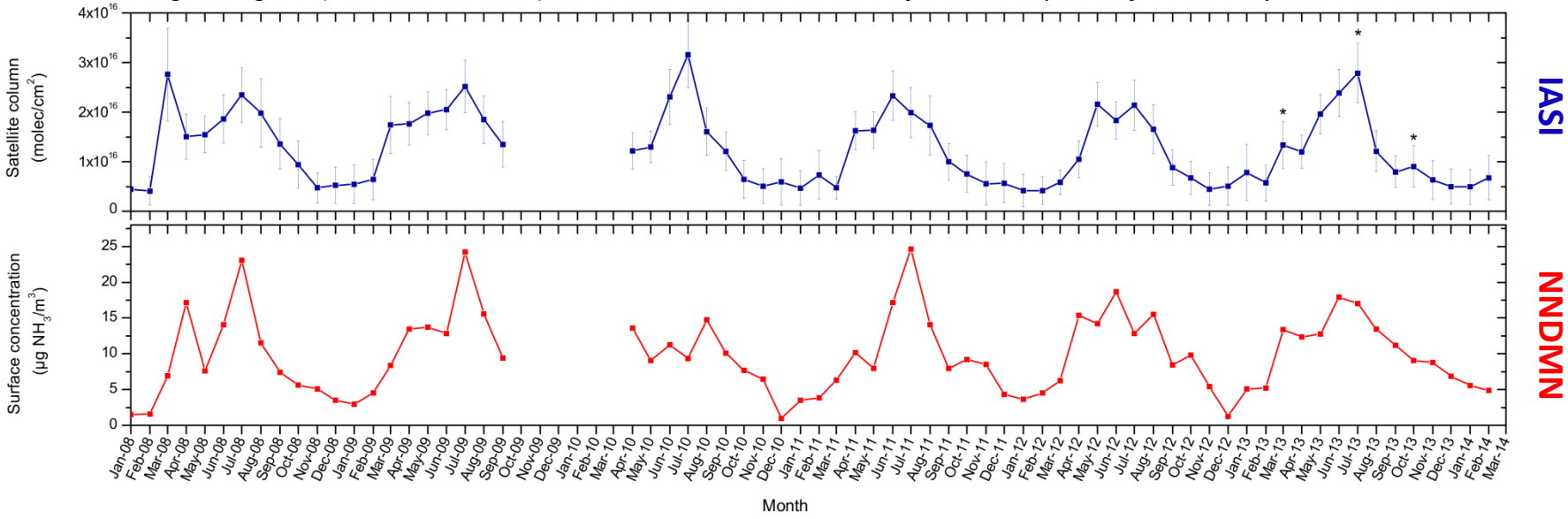
Qualitative comparison: IASI columns vs. surface abundances

→ Use LUT profile to convert IASI-columns to IASI-surface abundances

(Van Damme *et al.*, AMT 2015)

Shangzhuang site (40.14°N, 116.18°E)

* = *fertilization peaks of March, July and October 2013*



Geophysical analyses of NH_3 distributions & time-series

Method (2) From HRI to NH_3 column …using a neural network (NN)!

$$NH_3 \text{ column} = HRI \times f(T, T_{surf}, P, H_2O, \sigma, z_0, \varepsilon, \text{angle})$$

(Whitburn *et al.*, accepted in JGR-atm)

Advantages:

- ✓ **Full atmospheric state** is taken into account as there is no limit on the number of input parameters
- ✓ **Full uncertainty analysis** is achieved by perturbing the input parameters
- ✓ **Reduced bias**, NN approach allowing negative columns
- ✓ **Flexible NH_3 profiles:**

Cases	1	2	3	4	5	6	7	8	9
% deviation from NH_3 column based on land LUT (Van Damme *et al.*, 2014)	-53.4	-44.9	-32.2	-8.4	+11.8	0	+3	+14.9	+41.4
NH_3 profile shape (* in km)	A *=0.1	A *=0.5	A *=1	A *=2	A *=3	B	C *=0.5	C *=1	C *=2

- ➢ Study of fires
- ➢ Model comparisons
- ➢ In-situ measurements

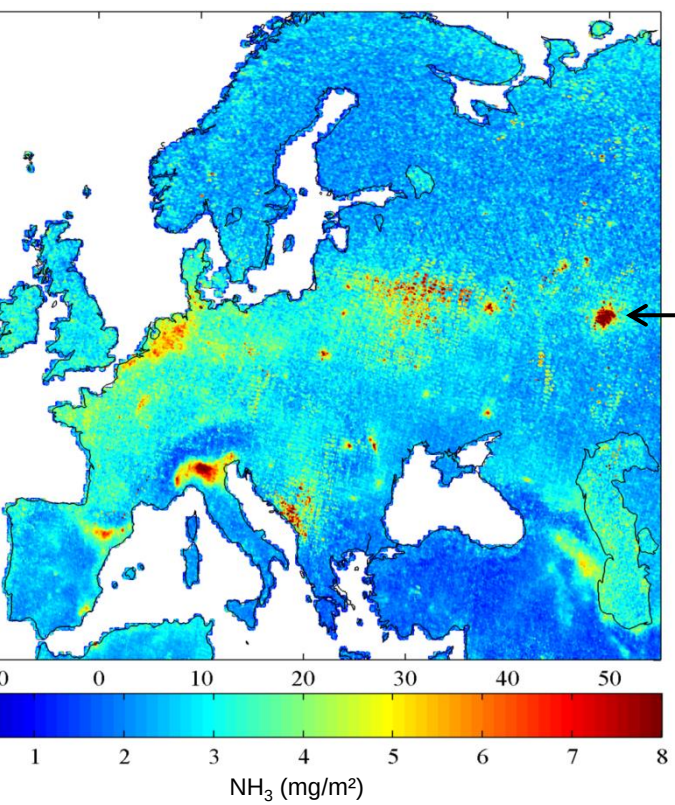
Geophysical analyses of NH_3 distributions & time-series

Model comparison

Inter-annual variability
4-yr comparison (2008→2011)

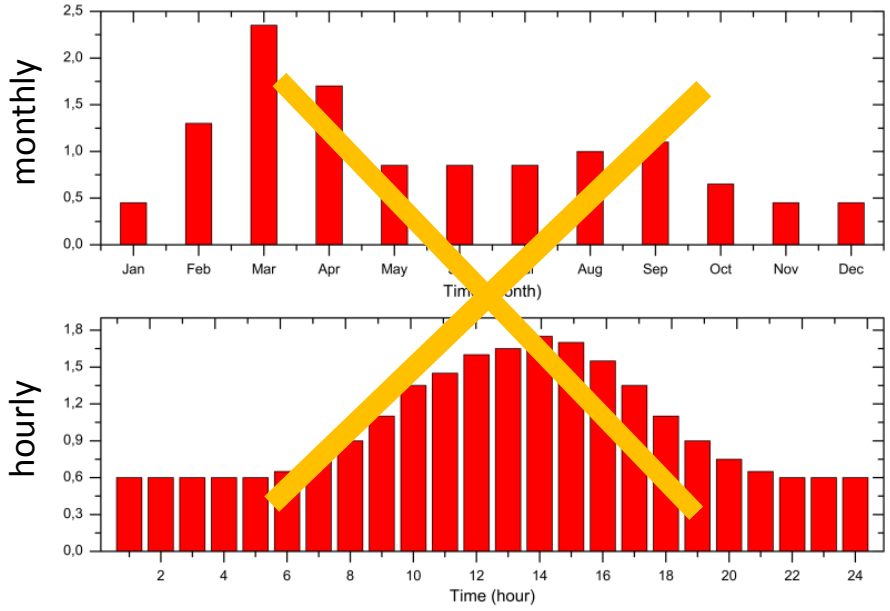
○ = industrial hot spots

LOTOS-EUROS (IASI) simulations | IASI observations | IASI error | IASI - LOTOS-EUROS



Intra-annual variability

Fixed temporal profile for agricultural emissions used for the entire European domain in the model



➢ First time that industrial NH_3 sources are obse… underestimation of emission in the inventories

➢ Limitations of the use of the same fixed temporal profile + added value of IASI-NH_3 data set

(Van Damme *et al.*, JGR-Atm 2014)