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Sensitivity of ammonia and particulate nitrate in Europe to dry deposition parameterization

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Ammonia

- gaseous ammonia plays an important role on the acidity of precipitation and formation of inorganic aerosols
- ammonia (NH_3) and ammonium (NH_4^+) are also nutrients (as fertilizers)

sources

agriculture

sinks

$$2NH_3 + H_2SO_4 \longrightarrow (NH_4)_2SO_4$$

$$NH_3 + HNO_3 \rightleftharpoons NH_4NO_3$$

dry and wet deposition

Impacts

acidification
eutrophication
toxicity to plants
loss of plant diversity

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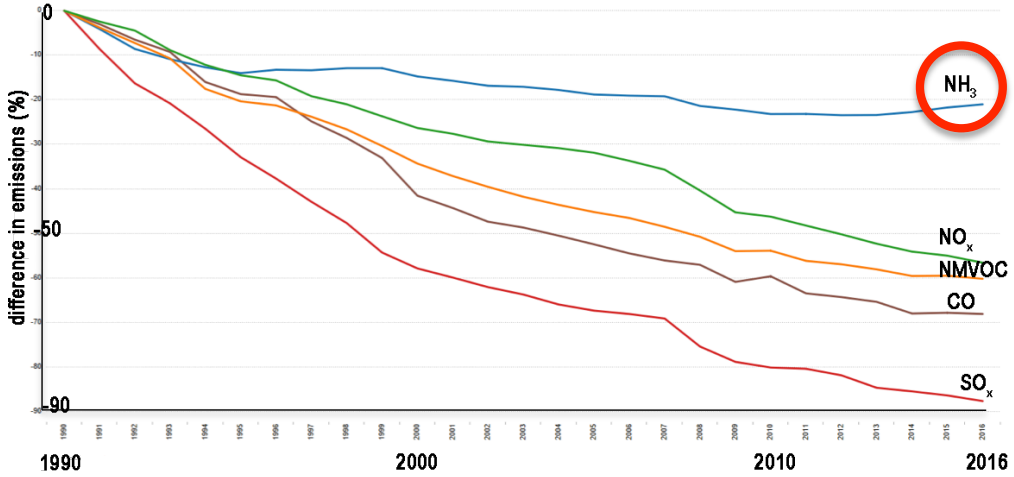
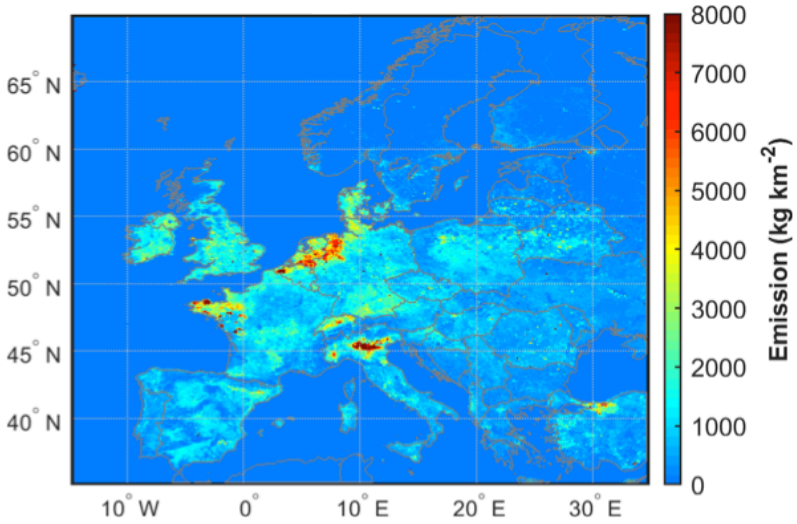
Emissions

Relative change in EU emissions 1990-2016



from EEA, 2018

NH_3 emissions in 2010 (TNO-MACCIII)



Anthropogenic emissions have been reduced substantially in Europe since 1990s, except ammonia

Highest ammonia emissions are around Benelux, north-west France and northern Italy

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Dry Deposition

resistance model for gases (Wesely, 1989)

$$V_d = \frac{1}{R_a + R_b + R_s}$$

where

V_d : deposition velocity (cm s^{-1})

R_a : aerodynamic resistance (s cm^{-1})

R_b : boundary resistance (s cm^{-1})

R_s : surface resistance (s cm^{-1})

R_s was replaced by canopy resistance (R_c) in Zhang model (Zhang et al. 2003)

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Goal

- In earlier CAMx versions surface resistance scaling parameter (R_{sc}) was set to zero for strong acids and ammonia given their strong rate of uptake by biota and other surfaces
- Starting with the version CAMx 6.50, the dry deposition parameter for ammonia was changed to more appropriately estimate NH_3 removal rates
- The parameter R_{sc} strongly influences NH_3 deposition velocity, affecting also the concentration of NH_3

How much the modeled ammonia and inorganic aerosol concentrations in Europe are affected due to the change in surface resistance parameter in CAMx?

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Method

- CAMx 6.50
- chemical mechanism: CB6r2
- simulation period: 2010
- horizontal resolution : 0.250^{o} x 0.125^{o}
- 14 layers, first layer ~20 m
- meteorology : WRF 3.7.1
- anthropogenic emissions :TNO-MACCIII
- biogenic VOC emissions : PSI-model
- boundary conditions: MOZART
- deposition: Zhang model

2 simulations

- R_{sc}= 1 (as in new version 6.50)
- R_{sc}= 0 (as in previous versions)

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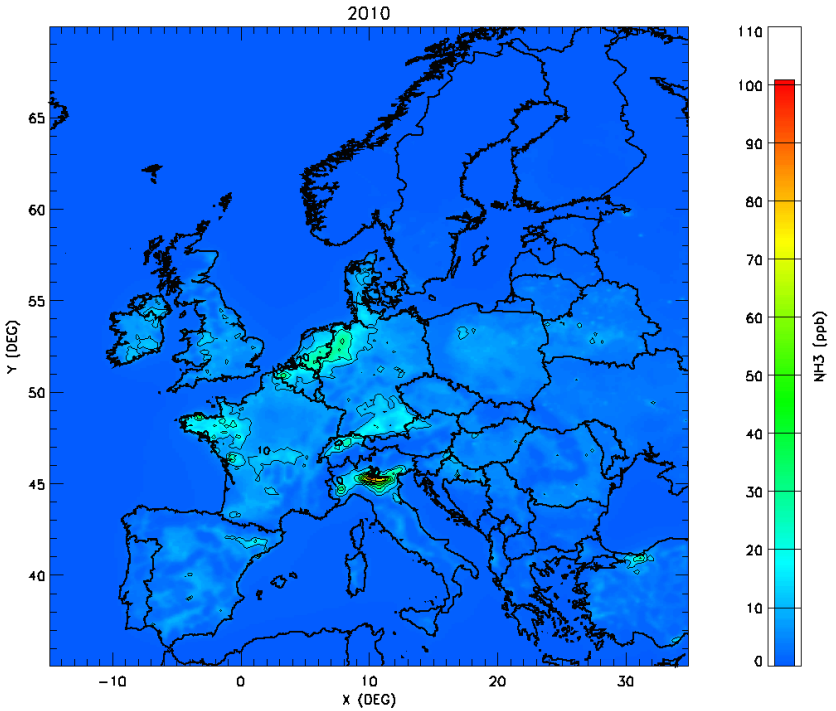
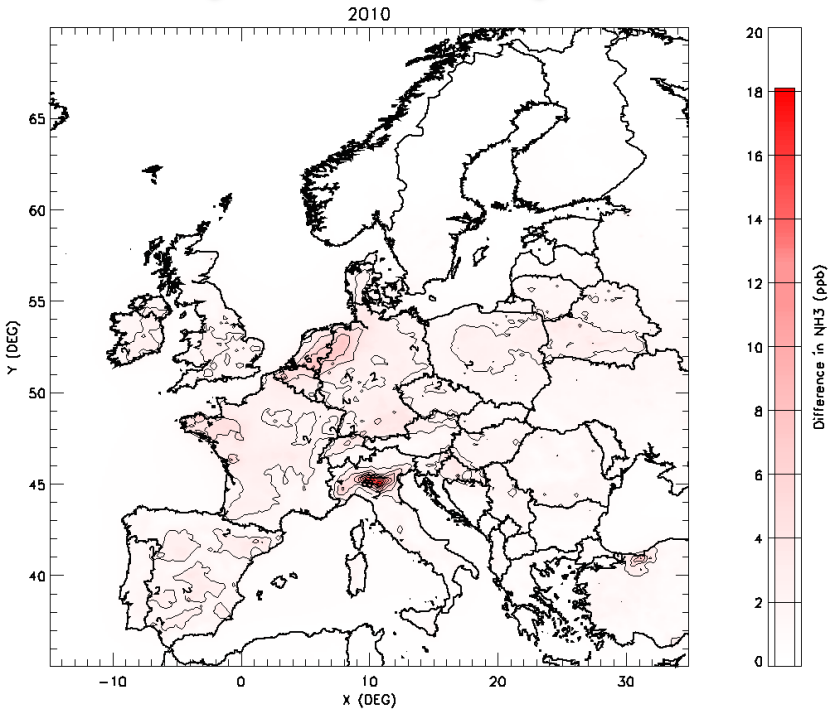
NH_3: Annual concentrations (ppb)

NH_3 (R_{sc}=1)



NH_3 (R_{sc}=1) – NH_3 (R_{sc}=0)



New R_{sc} parameter for ammonia in deposition velocity calculations led to an increase in NH_3 concentrations by about 30-50% over central Europe

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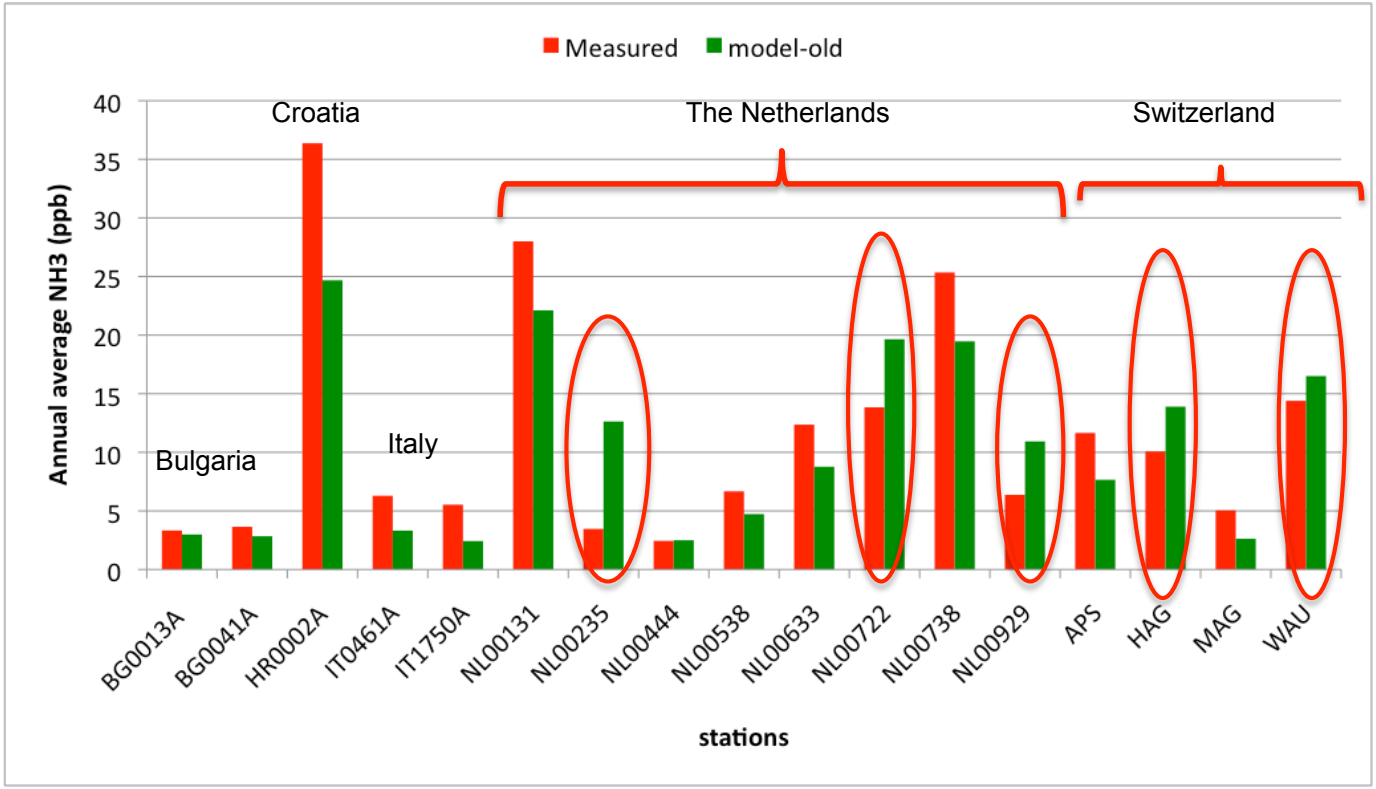
Comparison with measurements



Using the previous parameterization, ammonia was underestimated at most of the locations except a few sites

Measurements are from AirBase and FUB (CH)

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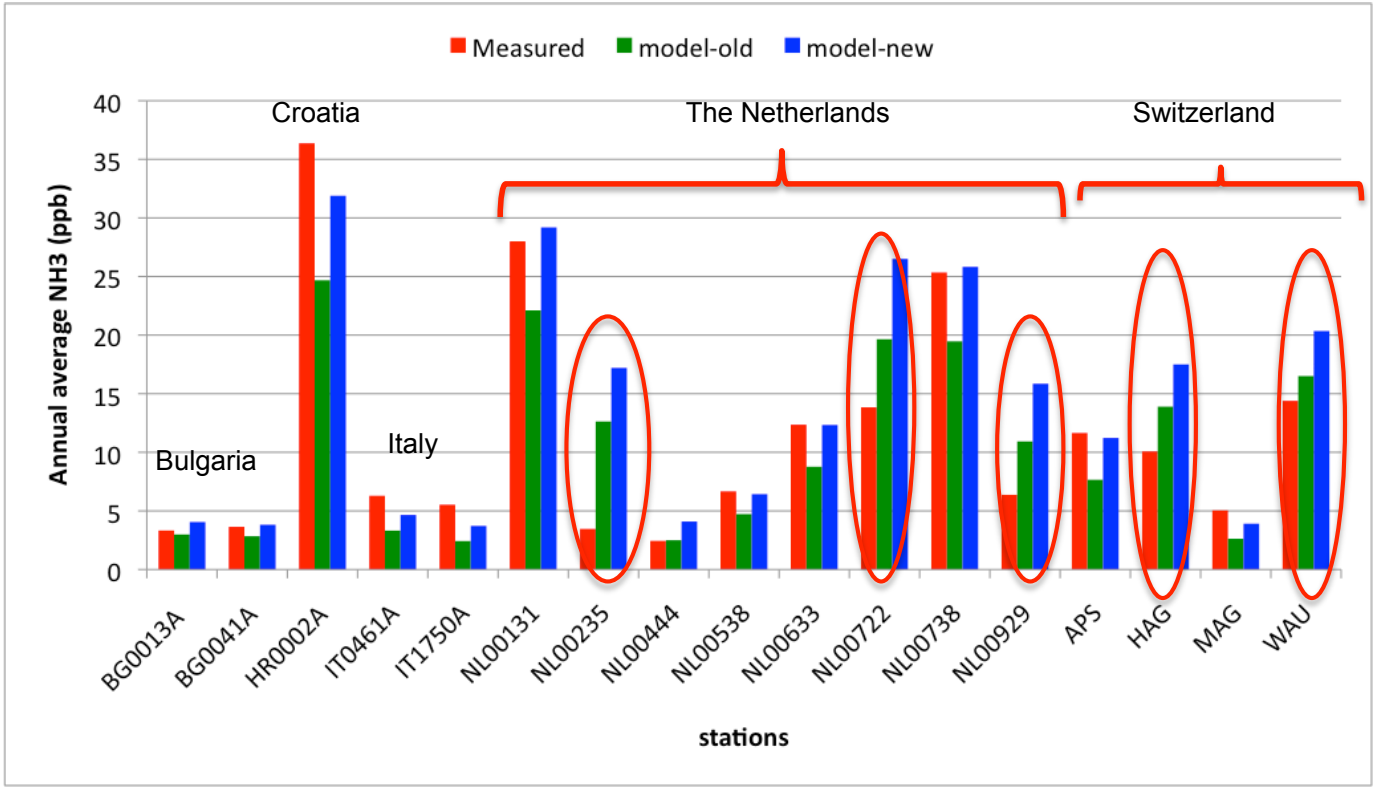
Comparison with measurements



The model performance for ammonia became better at most of the stations except a few sites where ammonia was even more overestimated than before

Measurements are from AirBase and FUB (CH)

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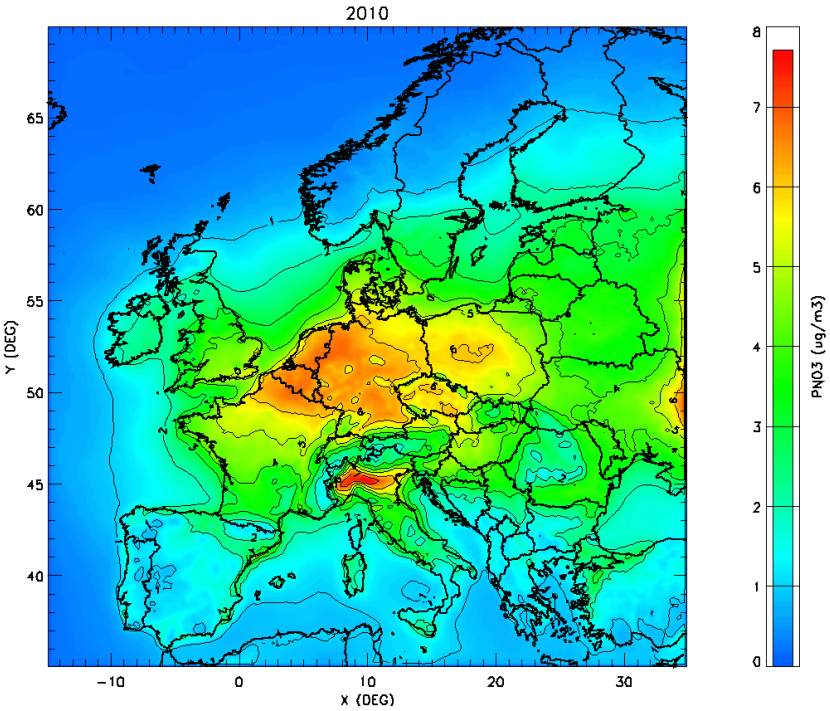
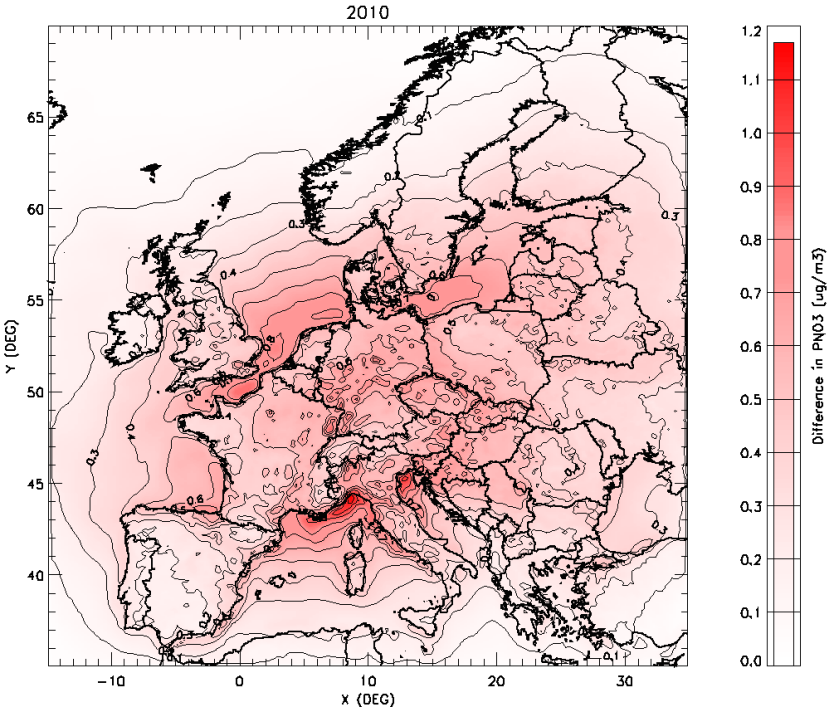
NO_3^- : Annual concentrations ($\mu g\ m^{-3}$)

NO_3^- (R_{sc}=1)



NO_3^- (R_{sc}=1) – NO_3^- (R_{sc}=0)



Increased ammonia concentrations led to an increase in particulate nitrate (NO_3^-) concentrations over central Europe (20%) and along the coastal regions (30%)

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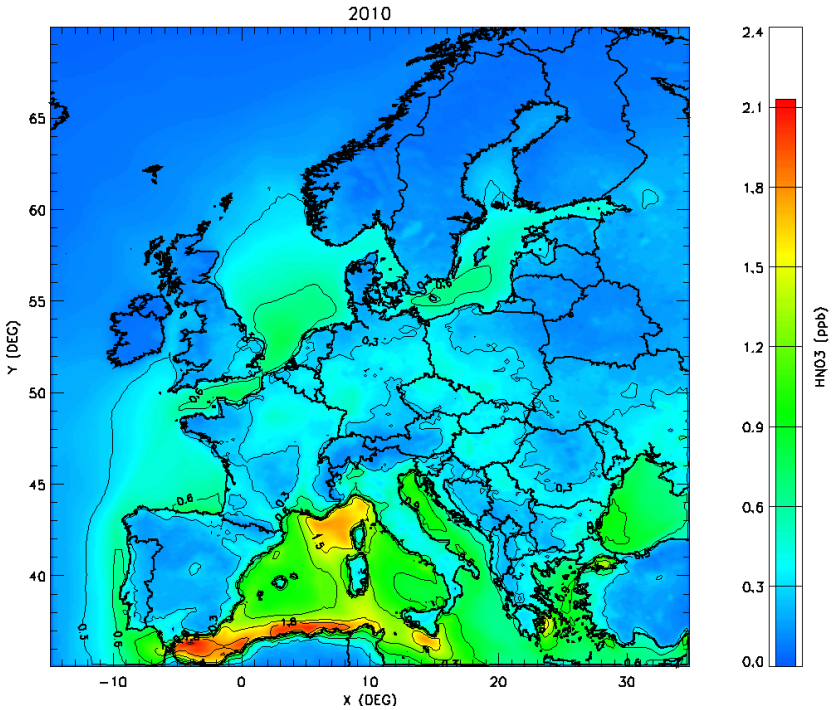
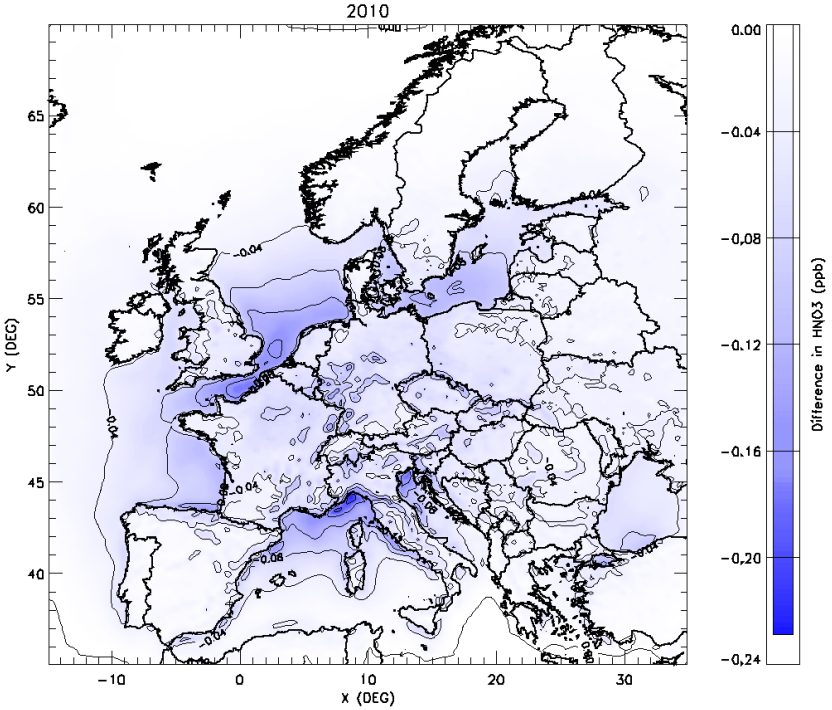
HNO_3 : Annual concentrations (ppb)

HNO_3 (R_{sc}=1)



HNO_3 (R_{sc}=1) – HNO_3 (R_{sc}=0)



HNO_3 concentrations decreased by about 20-30% because more ammonia reacted with HNO_3 increasing particulate nitrate (NO_3^-)

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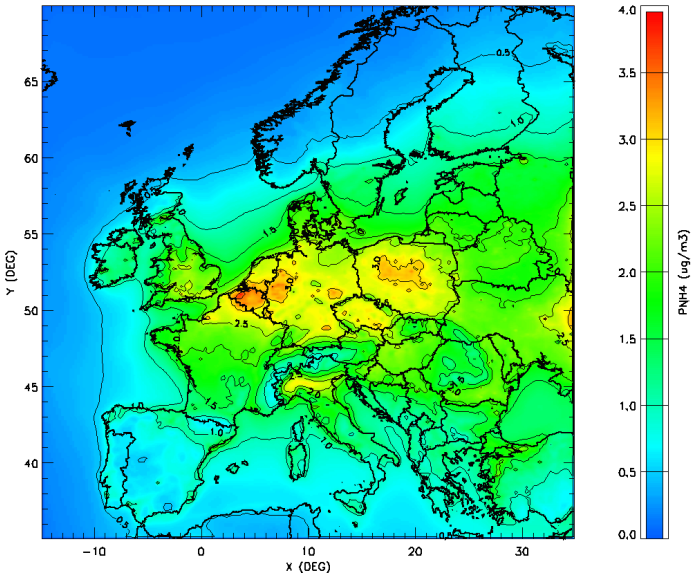
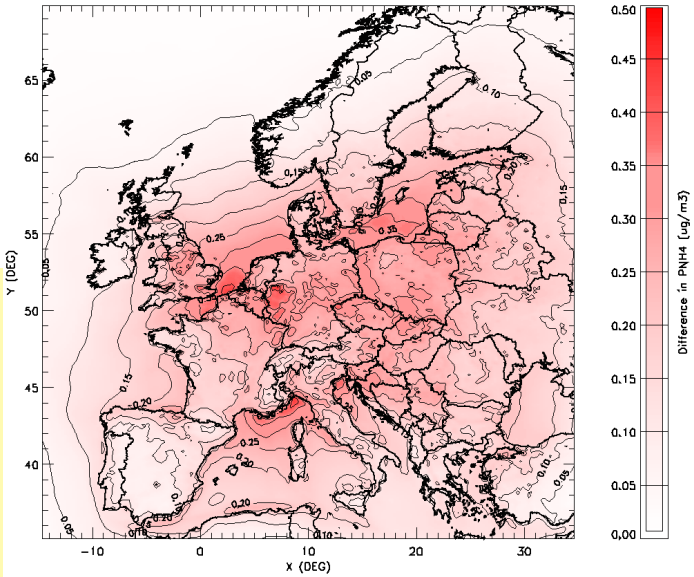
NH_4^+ and SO_4^{2-} ($\mu g\ m^{-3}$)

NH_4^+ (R_{sc}=1) 2010

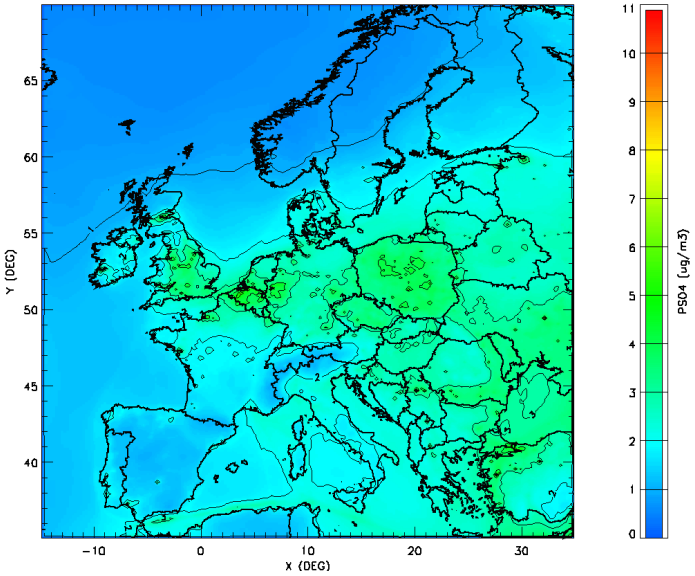


NH_4^+ (R_{sc}=1) – NH_4^+ (R_{sc}=0) 2010

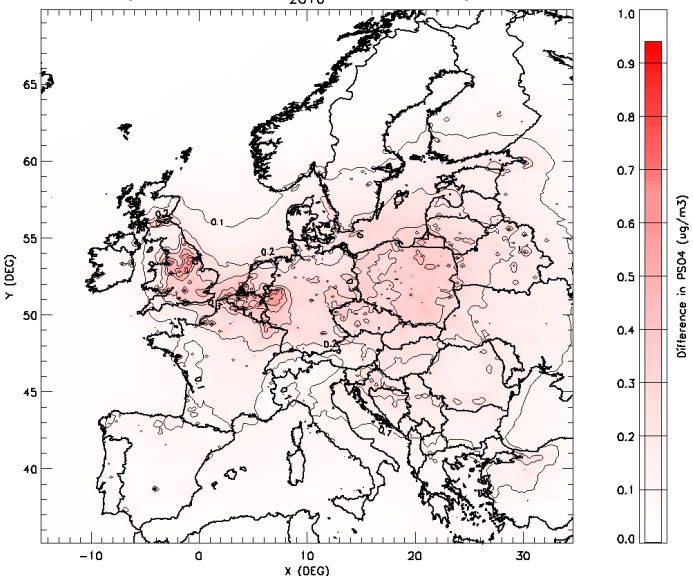


NH_4^+ and SO_4^{2-} increased by about 10%

SO_4^{2-} (R_{sc}=1) 2010



SO_4^{2-} (R_{sc}=1) – SO_4^{2-} (R_{sc}=0) 2010



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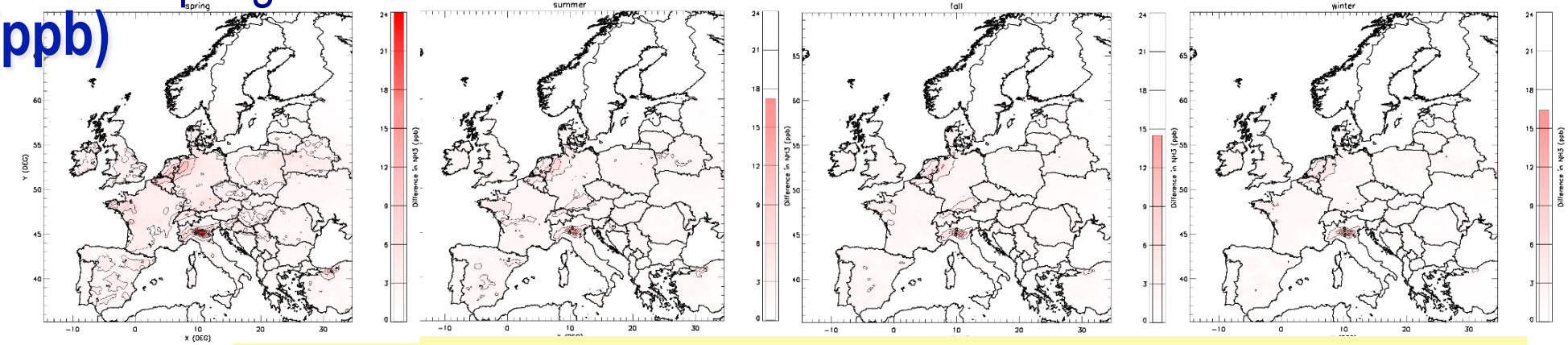
Change in seasonal concentrations

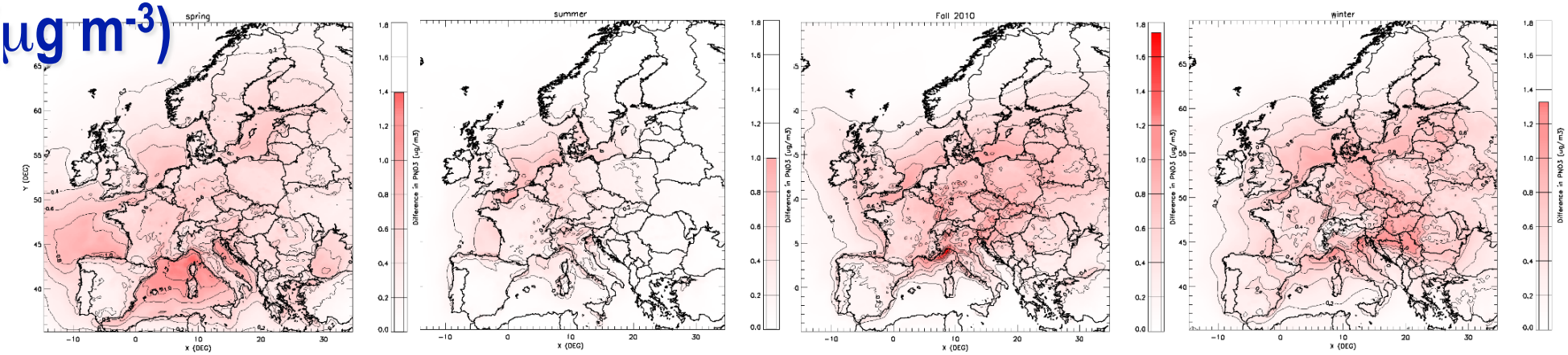
ΔNH_3 (ppb)

spring summer fall winter



NH_3 concentrations increased more in spring

ΔNO_3 ($\mu g\ m^{-3}$)



particulate nitrate concentrations increased more in spring (south, coast) and fall (north, land)

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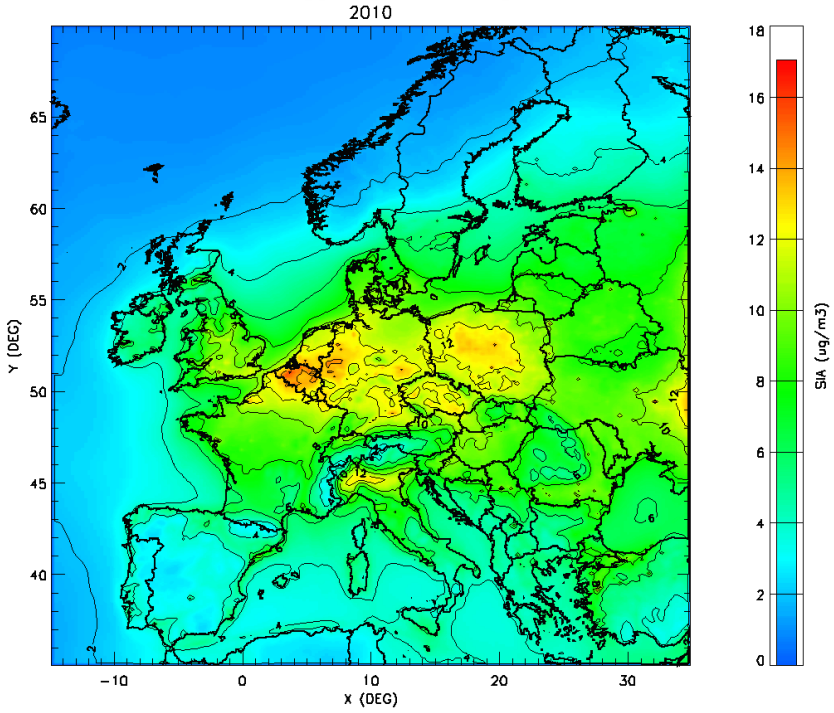
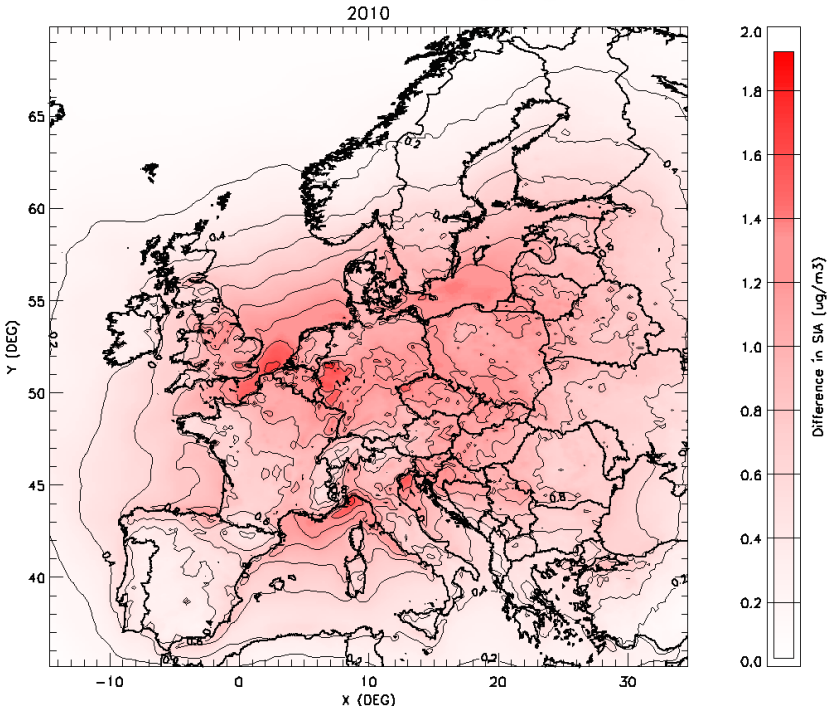
Secondary Inorganic Aerosols (SIA)

SIA ($\mu g\ m^{-3}$)



change in SIA ($\mu g\ m^{-3}$)



As a result of the change in dry deposition of ammonia, SIA concentrations increased by about 10% in central Europe and 20% along the coastal areas

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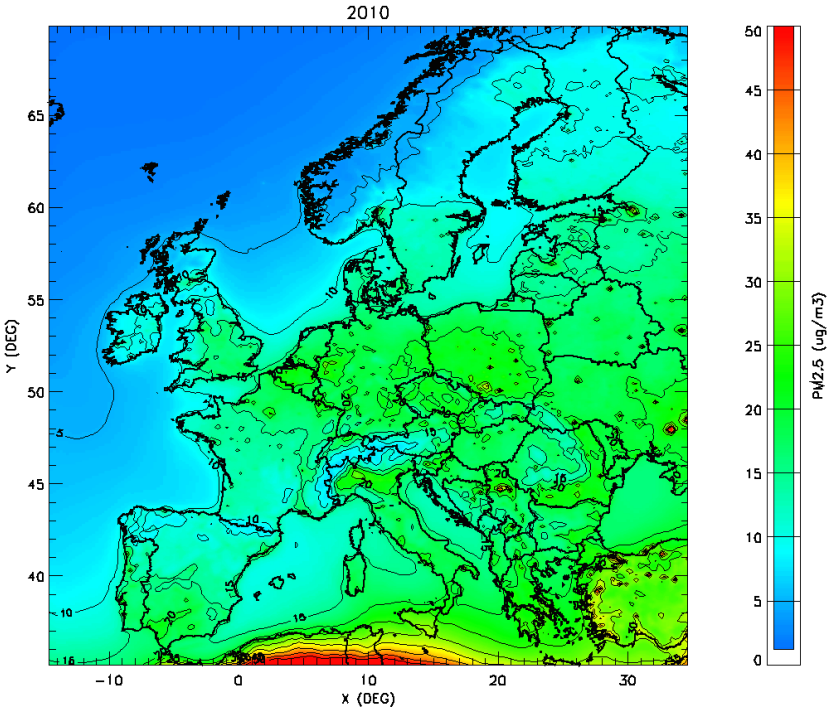
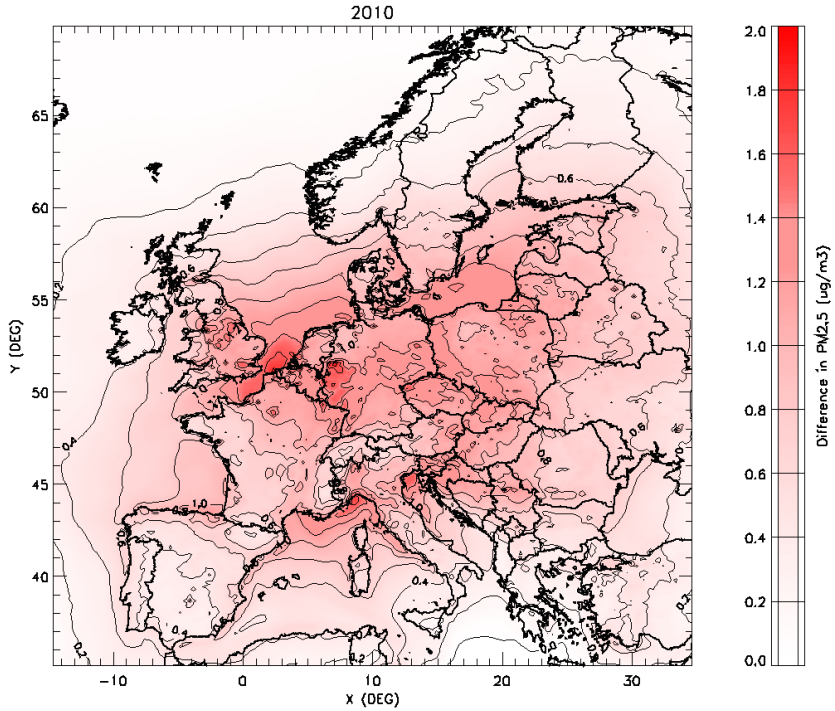
Annual $PM_{2.5}$ ($\mu g\ m^{-3}$)

$PM_{2.5}$ (R_{sc}=1)



$PM_{2.5}$ (R_{sc}=1) – $PM_{2.5}$ (R_{sc}=0)



The change in dry deposition of ammonia led to an increase in $PM_{2.5}$ concentrations by about 5-10% due to increased concentrations of secondary inorganic aerosols

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Conclusion

the change in the dry deposition parameter for ammonia in the latest CAMx version (6.50)

leads to an increase of 30-50% in annual ammonia concentrations in Europe, improving the model performance only at sites where ammonia was previously underestimated

effect

+30-50% on NH_3

↓

+20-30% on NO_3^-

↓

+10-20% on SIA

↓

+5-10% on PM2.5

- increased NH_3 leads to :
  - a decrease in HNO_3 concentrations
  - an increase in particulate nitrate by about 20% in central Europe and 30% along the coastal regions
- the total effect on PM2.5 is about +5% in central Europe, +10% along the coastal areas

more updates for NH_3 are expected soon in the next CAMx version

Thank you

Acknowledgements

TNO

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