National scale inversion of NO_X emissions in France constrained by TROPOMI NO_2 tropospheric columns

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LSCE Laboratoire des Sciences du Climat & de l'Environnement

ARGONAUT PollutAnts and Greenhouse Gases EmissiOns MoNitoring from SpAce at high ResolUTion

The recent progress in NO_2 measurement from space thanks to TROPOMI-S5P allows us to refine the scale at which we can monitor NO_X emissions. With regional chemistry-transport model CHIMERE operating at high resolution, we study how to assimilate NO_2 tropospheric columns to constrain bottom-up NO_X emissions inventories at 50 to 10 km and a 1-day resolution, moving from OMI to TROPOMI. Our analysis relies on the variational mode of the newly developed Community Inversion Framework CIF paired with the CTM and its adoint code.
The analysis indicates the difficulties to assimilate numerous satellite observations within a large dimension problem and to deal with the modeled-observed patterns misfits, due to CTM or weather errors, that arise when working at such resolution. We aim at developing an efficient emission monitoring tool for reactive species such as NO_X with image processing techniques to benefit from high resolution down to the regional scale.

NO_X variational inversions using CIF[1]-CHIMERE

Prior fluxes
- NO_X anthropogenic emissions from TNO-GHGco-v3 [1.] or CITEPA/INERIS INS [2.]
- NO_X biogenic emissions from MEGAN

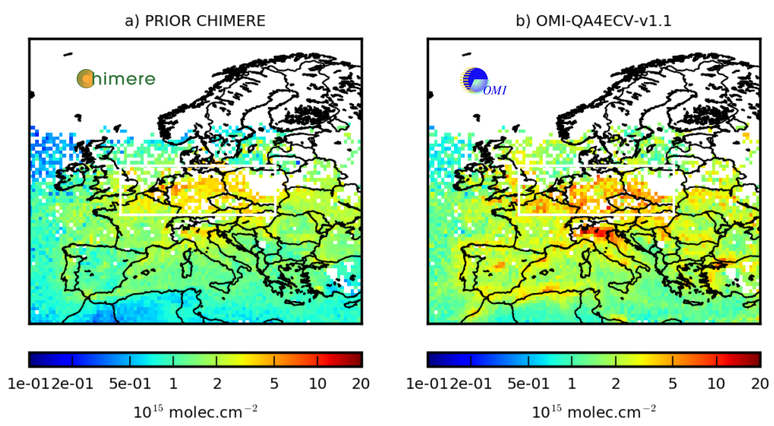
Model
- Regional chemistry-transport model CHIMERE at 0.5°x 0.5°x 17 vertical levels [1.] or 0.1°x 0.1°x 20 vertical levels [2.]
- MELCHIOR-2 module for gaseous chemistry
- Adjoint of CHIMERE including adjoint of chemistry
- ECMWF meteorological fields

Observations
- Satellite retrievals of NO_2 from OMI-QA4ECV-v1.1 [1.]
- TROPOMI-S5P [2.]

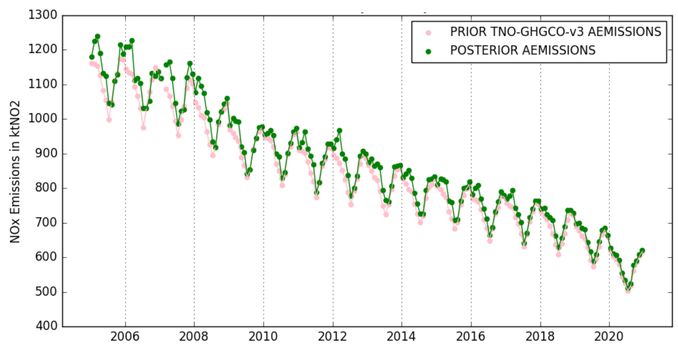
[1]Community Inversion Framework, (CIF; Berchet et al., 2021, GMD)
[1.] Europe, [2.] France

1. Inversion for European NO_x emissions at 0.5° with OMI

- Strong underestimation of the NO2 simulated TVCDs compared to OMI-QA4ECV observations, seen for all seasons:
  → underestimation of prior emissions?
  → biases in the observations?
- Consistent with the literature [e.g. Huijnen et al., 2010; Miyazaki et al., 2017; Visser et al., 2019]



Monthly mean of NO_2 tropospheric columns in January 2020 (in 10^{15} molec.cm^{-2})

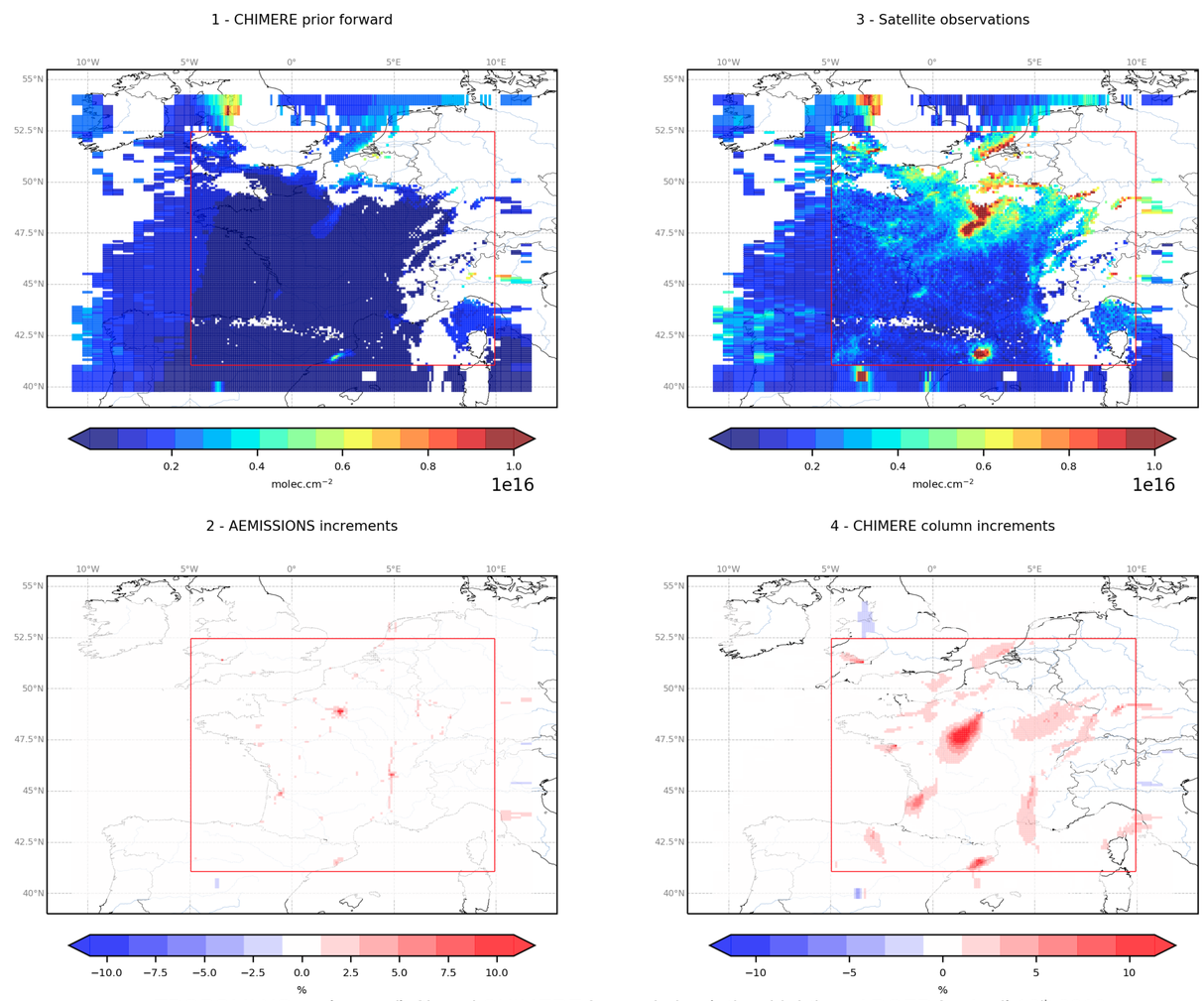


Monthly prior and posterior estimates of the NO_x anthropogenic emissions from 2005 to 2020 over continental land (in $ktNO_2$)

- The posterior NO_x emissions are slightly changed compared to the prior ones during winter mainly because of a lack of observations
- The inversion **mainly applies positive increments to the prior anthropogenic emissions in spring and in summer**

2. Inversion for French NO_x emissions at 0.1°

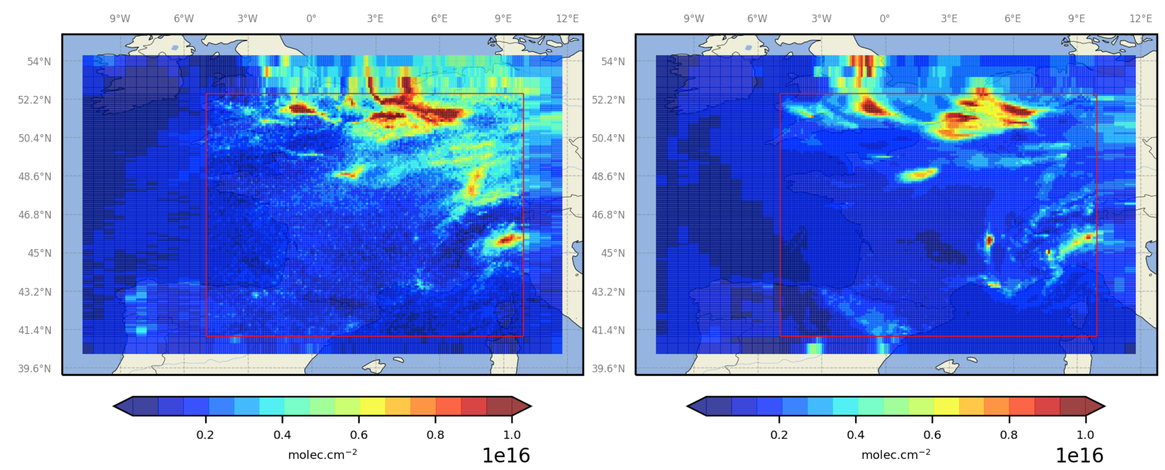
- Will working at high resolution change the correction magnitude?
- The bottum-up inventory seems to underestimate anthropogenic fluxes when comparing the CHIMERE forward simulation (1) with TROPOMI observations (3)
- The assimilation system compute spatially coherent (positive) corrections regarding the difference between the *prior* and observations (2)
- But the positive flux increments (up to +10%) are not sufficient to match TROPOMI in the *a posteriori* forward simulation (4)
- As seen at European scale, the computed corrections are relatively limited



TROPOMI NO_2 columns (left) and CHIMERE forward simulation (right) on ARGFR for April 12th 2019

3. Studying methods to help working with NO_X at high resolution

- High resolution highlights shape and position misfits between observed and simulated patterns due to the meteorological/transport errors (see figure below)
- We aim at generalizing detection and inversion techniques applied to plumes/patterns associated with individual point sources that are today tested for individual metropolitan plumes or thermal power plants)
- The goal is to process the non diffuse part of the signal on which transport errors have a consequent weight



TROPOMI NO_2 columns (left) and CHIMERE forward simulation (right) on ARGFR for March 3rd 2020

Current issues and next steps

- Studying the consistency between OMI and TROPOMI
- Define and use super-observations methods to assimilate TROPOMI columns
- Small corrections applied by the assimilation system: analyse the impact of highly reactive species inversion problem's non-linearity?
- 30%-50% prior uncertainties → too low at national scale? Without spatial correlations → leads to local corrections without national impact?