# SPECIAL PROJECT PROGRESS REPORT

All the following mandatory information needs to be provided. The length should *reflect the complexity and duration* of the project.

| | |
|---|---|
| **Reporting year** | ……2020……………………………………………………… …… |
| **Project Title:** | … Top-down estimate of chlorofluorocarbon emissions in Europe using a mesoscale inverse modeling technique |
| **Computer Project Account:** | spitgraz.……………………………………………………… ……… |
| **Principal Investigator(s):** | Francesco Graziosi …………………………………………………………… |
| **Affiliation:** | … Institute of Atmospheric Sciences and Climate - National Research Council (ISAC-CNR)…………… |
| **Name of ECMWF scientist(s) collaborating to the project** (if applicable) | …………………………………………………………… …………………………………………………………… |
| **Start date of the project:** | 17/12/2018………………………………………… |
| **Expected end date:** | 31/12/2020………………………………………… |

## Computer resources allocated/used for the current year and the previous one

(if applicable)

Please answer for all project resources

| | | Previous year | | Current year | |
|---|---|---|---|---|---|
| | | Allocated | Used | Allocated | Used |
| **High Performance Computing Facility** | (units) | 600000 | 0 | 600000 | 202871.08 |
| **Data storage capacity** | (Gbytes) | 20000 | 0 | 25000 | 18000 |

## **Summary of project objectives** (10 lines max)

The main goals are :
European emissions estimation of main Chlorofluorocarbons (CFCs), CFC-11, CFC-12,
Estimate the trend of emissions from 2008 to 2019, of different species ($CCl_4$, $CH_3CCl_3$ and HCFC-22).
Allocate the source distribution over the European domain.
Increasing understating of type of sources, by product, banks o direct emissions.
Estimate the pro capita emissions of single countries.
Estimate the season emission cycles…………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………….

## **Summary of problems encountered** (10 lines max)

Compilation and running the dispersion model on machine…………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………...

## **Summary of plans for the continuation of the project** (10 lines max)

Increasing the model simulation resolution to get results that are more accurate. Estimate the emissions season cycle of species studied. Improving the understand of sources nature of species analysed.
…………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………………….……

## **List of publications/reports from the project with complete references**

Oral presentation : "Estimating CFC-11 emissions over Western Europe from atmospheric observations" at CFC-11 Symposium 25-27 March 2019, Vienna, Austria

## **Summary of results**

If submitted **during the first project year**, please summarise the results achieved during the period from the project start to June of the current year. A few paragraphs might be sufficient. If submitted **during the second project year**, this summary should be more detailed and cover the period from the project start. The length, at most 8 pages, should reflect the complexity of the project. Alternatively, it could be replaced by a short summary plus an existing scientific report on the project attached to this document. If submitted **during the third project year**, please summarise the results achieved during the period from July of the previous year to June of the current year. A few paragraphs might be sufficient.

We run FLEXPART model in backward simulations with four receptors (Mt. Cimone (CMN), Mace Head (MHD), Jungfraujoch (JFJ) and Tacolneston (TAC)) over global domain, from 2008 to 2019. We estimated the emissions of CFC-11, CFC-12 $CH_3CCl_3$ $CCl_4$ and HCFC-22 for North Western Europe (NWEU) (Ireland, UK, France, Germany, Belgium, Luxembourg, The Netherlands, Denmark, Italy, Switzerland, Austrial) using atmospheric inverse modelling. With the aim to evaluate the model performance, we run several sensitivity tests. Briefly, 1) we performed the inversion using two different kind of a priori fields, a uniform a prior (named flat) and population distribution fields (named pop); 2) we test different stations geometry (this test is shown for CFC-11 only). The tests highlighted nice model stability, with an average value of less than 22% of emissions variability between different model settings for all species.
Fig 1 shows the average yearly geographical sensitivities to surface emissions (footprints) from the four observation sites obtained from our simulations for the period 2013-2019. Based on this information, emissions from NWEU were considered sufficiently well resolved to be reported.

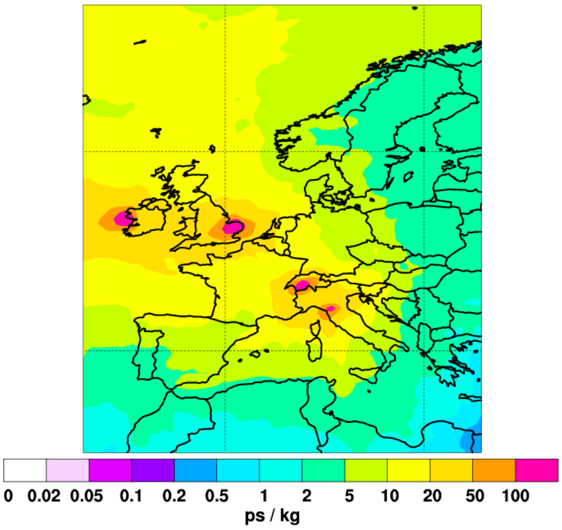



Fig 1 Footprint emission sensitivity in picoseconds per kilogram (ps $kg^{-1}$) obtained from FLEXPART 20 d backward calculations averaged over all model calculations over two years (Jan 2013- Dec 20019), using CMN, JFJ, TAC and MHD receptors.

Our preliminary inversion results show NWEU emissions of CFC-11, CFC-12, $CH_3CCl_3$, $CCl_4$ and HCFC-22 declined from 2008 to 2019. From 2008 onwards information from three observation stations is available and all four inverse systems have been used to estimate WEU CFC-11 emissions. With the introduction of the station at TAC in 2012, estimates using data from four stations are possible. The yearly CFC-11 CFC-12 $CH_3CCl_3$ $CCl_4$ and HCFC-22 emissions from NWEU from the four inverse systems are shown in **Errore. L'origine riferimento non è stata trovata.**2. Panels a and b of Fig 2 shows CFC-11 emissions using three stations (2008-2012) and four stations (2013-2019) respectively. Panels from c to f of Fig 2 shows CFC-12, $CH_3CCl_3$ $CCl_4$ and HCFC-22 yearly emissions from 2008 to 2019.

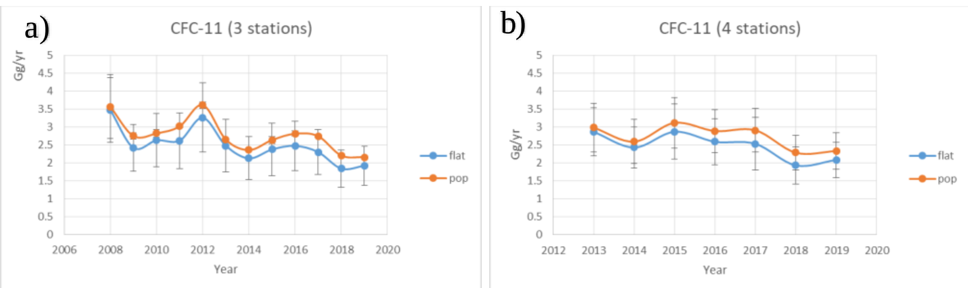



**Fig 2** Graphs showing the NWEU emissions (Gg) over the period 2008-2019. The species analyzed are reported on the graph title, for CFC-11 (using 3 and 4 stations) (panel a, b respectively) and CFC-12 (panel c) we calculate a posteriori emission using flat and pop a priori filed (blue and orange line), meanwhile, for CH3CCl3, CCl4 and HCFC-22 (panel d, e, and f respectively) we reported the inversion obtained using flat a priori emission (blue line).

The inversions from 2008 – 2019 shows an average yearly CFC-11 emission from NWEU of 2.49 ± 0.70 Gg using the flat prior and 2.77 ± 0.75 Gg using the population prior. The inversions using either prior show a decline in emissions across the time-series of 0.09 ($r^2$ = 0.6) and 0.08 ($r^2$ = 0.5) Gg $yr^{-1}$ respectively, this would equate to an annual decline in the bank of 4.7% (3 – 6%) and 4.3% (3 – 6%) respectively. The four station inversions from 2013 – 2019 using a flat prior show a linear yearly CFC-11 emission from NWEU of 2.4 ± 0.60 Gg, with a linear rate of decline of 0.13 ($r^2$ = 0.62) Gg $yr^{-1}$, indicating an annual decline of 5.8% (4 - 8%). For the population prior the estimates are 2.71 ± 0.62 Gg, 0.10 ($r^2$ = 0.42) Gg $yr^{-1}$ and 5.3% (4 – 7%) respectively. These results demonstrate that the 3 and 4 station inversions are very consistent in the overall CFC-11 emission from NWEU. The year to year variability between the two setups all fall within the estimated uncertainty for each year, it is also clear that the addition of the TAC data adds uncertainty to the trend. In summary, the results indicate that NWEU has emitted on average ~2.4 Gg $yr^{-1}$ over the last decade and there has been an annual decline in emission of ~5% (3 – 8%). The CFC-12 average emission values 2008-2019 are 1.3 ± 0.40, 1.7± 0.47 Gg using flat and pop a priori emission field respectively. The $CH_3CCl_3$, $CCl_4$ and HCFC-22 average emission fluxes from 2008-2019 are 0.029 ± 0.09, 0.93 ± 0.30, 4.1 ± 1.3 Gg respectively.
The CFC-11 and CFC-12 average annual geographical distribution estimated by the inverse systems for 2008-2019 and a flat and pop prior are shown in Fig 2. The models exhibit strong consistency in the distribution of emissions for both gasses, all systems point to elevate (compared to the other regions) emissions over Belgium and southern Netherlands. A more precise location is not possible given the sparsity and the precision of the observation network and the uncertainty of the atmospheric modelling. The main emissions area for $CCl_4$ and $CH_3CCl_3$ is localized south of France as shown in Fig 4, meanwhile the HCFC-22 emission pattern is more sparse over the domain. These different emission distributions suggest a different emission sources, even if common hot spot emission region is retrieved on Netherland and Benelux areas for all the species. Further investigation is needed to understand the nature of the common sources region.

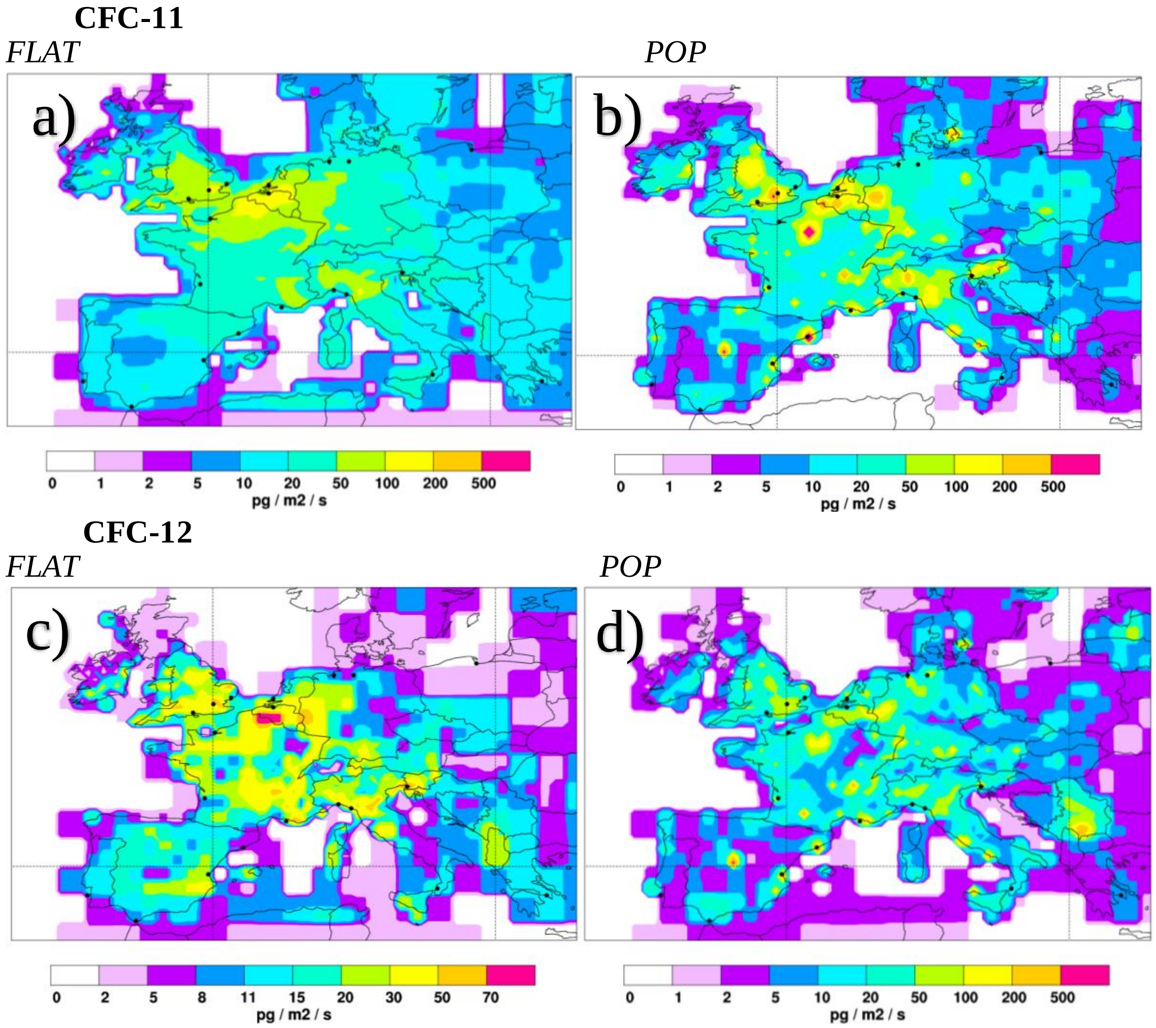
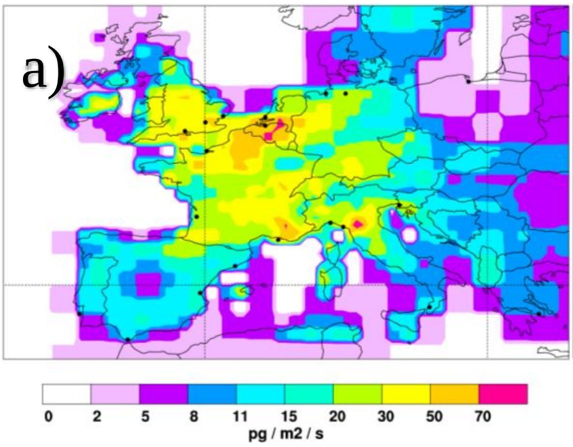
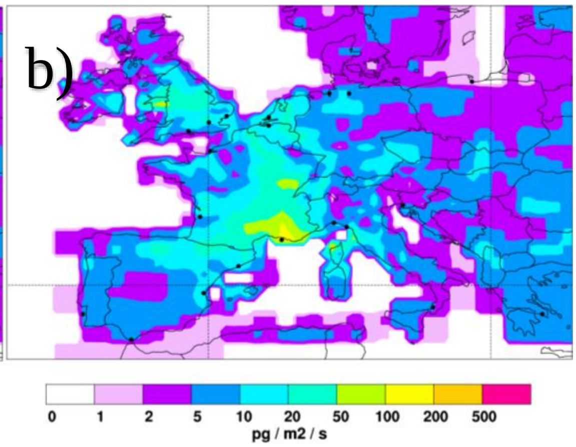



Fig 3 Average 2008-2019 a posterior emission fluxes distribution for CFC-11, CFC-12, using flat and pop (a, b, c, d and e panel) a priori emission field

**CH3CCl3**
*FLAT*




**CCl4**
*FLAT*




**HCFC-22**
*FLAT*

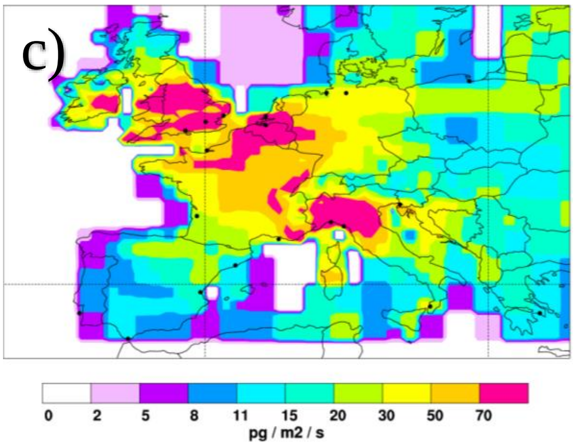



Fig 4 Average 2008-2019 a posterior emission fluxes distribution for CH3CCl3 CCL4 and HCFC-22 (a, b, c panel) using flat a priori emission field.

Due to the limited number of computer resource available for the project part of the simulations were conducted on local machine.

……………………………………………………………………………………………………………
……………………………………………………………………………………………………………
……………………………………………………………………………………………………………
……………………………………………………………………………………………………………
……………………………………………………………………………………………………………
……………………………………………………………………………………………………………
…………………………………………