SPECIAL PROJECT FINAL REPORT

All the following mandatory information needs to be provided.

Project Title:	Top-down estimate of chlorofluorocarbon emissions in Europe using a mesoscale inverse modelling technique
Computer Project Account:	spitgraz…
Start Year - End Year :	2018- 2020.
Principal Investigator(s)	Francesco Graziosi
Affiliation/Address:	Institute of Atmospheric Sciences and Climate - National Research Council (ISAC-CNR)
Other Researchers (Name/Affiliation):	Federico Fierli (European Organisation for the Exploitation of Meteorological Satellites-EUMETSAT), Francesco Cairo (ISAC-CNR), Paolo Cristofanelli (ISACCNR)

The following should cover the entire project duration.

Summary of project objectives

(10 lines max)

The main goals are : West European emissions estimation of main Chlorofluorocarbons (CFCs), trichlorofluoromethane (CFC-11) dichlorodifluoromethane (CFC-12) and Carbon tetrachloride (CCl4). Estimate the trend of emissions from 2008 to 2020, and allocate the source distributions over the study domain. Increasing understating of type of sources, by product, banks o direct emissions. Estimate the pro capita emissions of single countries.…………………………
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Summary of problems encountered

(If you encountered any problems of a more technical nature, please describe them here.)

During the first six months, we had a problem in compiling both WRF-ARW and FLEXPART models. Additional problems were related to the compilation of inversion system.
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Experience with the Special Project framework

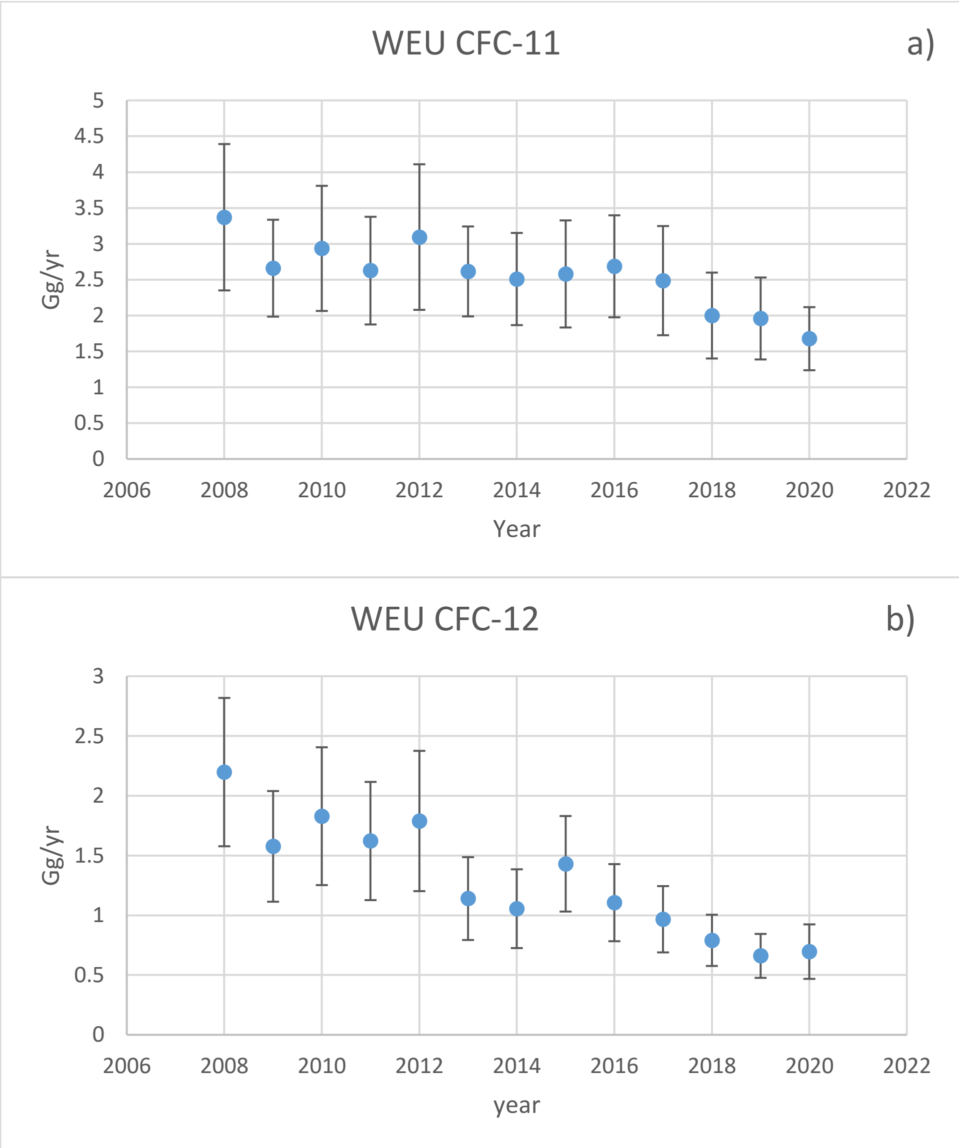
(Please let us know about your experience with administrative aspects like the application procedure, progress reporting etc.)

We don't have a negative remarks and we have found the Special Project framework nice, and well structured.………………………………………………………………………………

Summary of results

(This section should comprise up to 10 pages, reflecting the complexity and duration of the project, and can be replaced by a short summary plus an existing scientific report on the project.)

The aim of this project is to estimate the atmospheric emissions of trichlorofluoromethane (CFC-11) dichlorodifluoromethane (CFC-12) and Carbon tetrachloride (CCl4) from Western Europe (WEU) through a top down approach. CFC-11 CFC-12 and CCl4 emissions into the atmosphere are stratospheric ozone depleting substances and strong greenhouse gases. For these reasons, their production, consumption and use are controlled through the Montreal Protocol and amendments. For non-Annex 5 parties, including all of the countries in Western Europe, their production, consumption and use have been banned since 1995, for Annex-5 (developing) countries since 2010. However, a recent work [1] showed a slow-down in the global decline of atmospheric CFC-11 from 2013 due to an increase in global emissions of CFC-11 from EasternAsia. We investigate the potential contribution of WEU to global emissions. For this, we applied a top down approach, based on an atmospheric dispersion model in conjunction with observation data and Bayesian inversion technique. We use the in-situ high-frequency observations from four atmospheric monitoring stations; Mace Head (MHD) on the west-coast of Ireland, Tacolneston (TAC) in south-east UK, Jungfraujoch (JFJ) in the Swiss Alps and Monte Cimone (CMN) in the northern Apennine mountains in Italy. MHD, JFJ and CMN are part of the Advanced Global Atmospheric Gases Experiment(AGAGE) network, meanwhile TAC is part of the UK DECC network but is closely associated with AGAGE. All four share common calibration and are routinely inter-compared. The MHD, JFJ and CMN data are available from the year 2008 to 2020, meanwhile TAC observations start from the year 2013. The inversion approach is based on backward simulations with the LPDM FLEXPART [2] see also (http://transport.nilu.no/flexpart). The model was driven with operational analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) with 1 ◦×1 ◦ resolution. Every 3-h interval, 50 000 particles were released at the measurement point and followed backward in time for 20 d to calculate an emission sensitivity. The virtual particles were simulated without the removal process. We run the inversions from 2008 to 2020 for all of these atmospheric species. The estimation of gridded emissions is based on the analytic inversion method of [3] and expanded by [4], were all mathematical details can be found. We estimated the emissions of CFC-11, CFC-12 and CCl4 for WEU (Ireland, UK, France, Germany, Belgium, Luxembourg, The Netherlands, Denmark, Italy, Switzerland, Austria). With the aim to evaluate the model performances, we run several sensitivity tests. Briefly, 1) we performed the inversions using two different kinds of a priori fields, a uniform a prior (named flat) and population distribution fields (named pop); 2) we tested different station geometry. The tests highlighted nice model stability, with an average value of less than 22% of emissions variability between different model settings for all species. These tests were performed during the previous years, for this reason they are not reported in this report. Based on this information, emissions from WEU were considered sufficiently well resolved to be reported. In this report we show the emission obtained with a flat a priori emission field, viewed as a reference to a prior field. Here we report the inversion results from 2008 to 2020. The inversions from 2008 to 2012 are performed using 3 receptors, CMN JFJ and MHD receptors, meanwhile emission reported from 2013 to 2020 are obtained using all the monitoring sites.



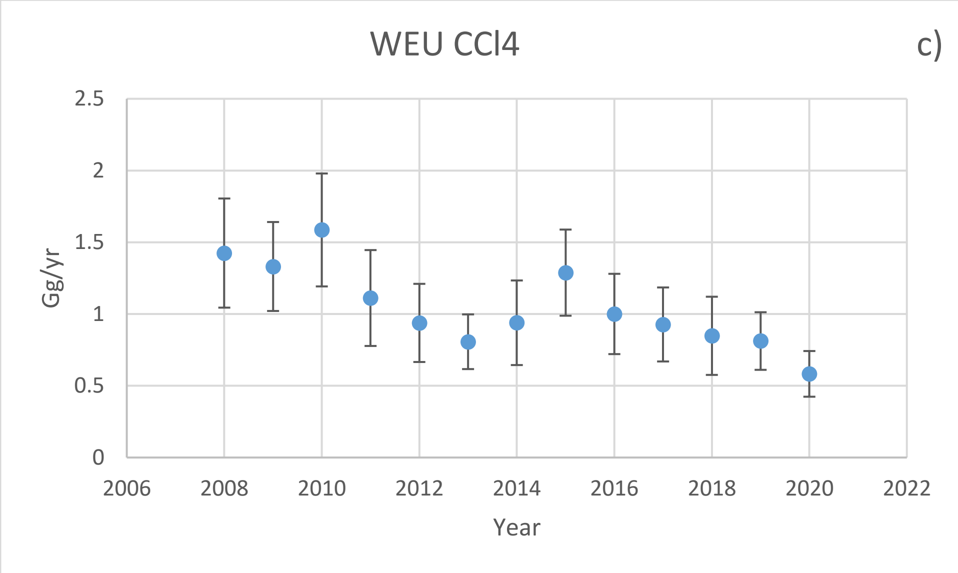
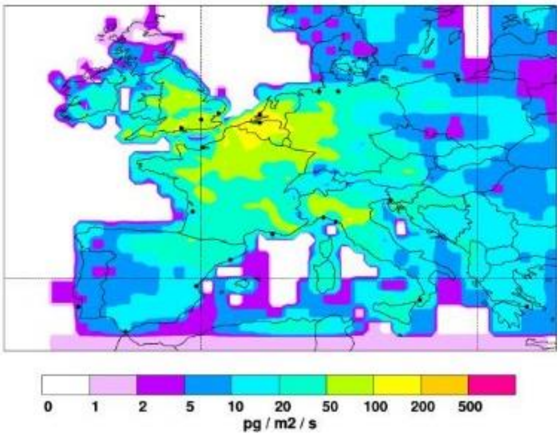
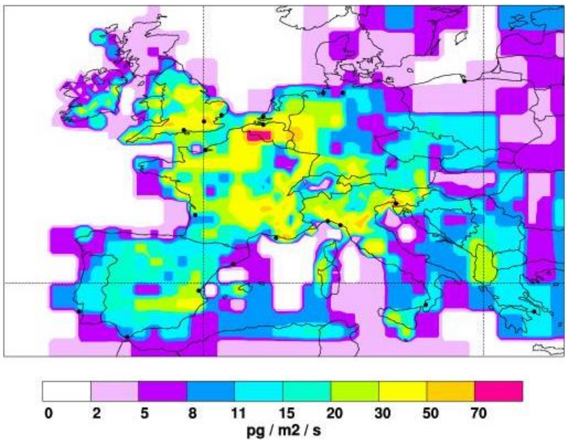


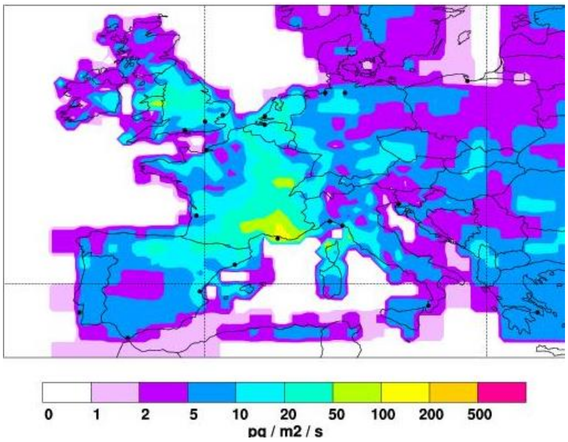
Fig 1 *Graphs showing the WEU emissions (Gg) over the period 2008-2020. The species analyzed are reported on the graph titles, CFC-11 (panel a),CFC12 (panel b)and CCl4 (panel c)*



a)



b)



c)

Fig 2 *Maps a, b and c of average emissions distribution (pg/m2/s) of CFC-11, CFC-12 and CCl4 respectively, from WEU area and over the period 2008-2020.*

The inversions from 2008 – 2020 shows an average yearly CFC-11 emission from WEU of 2.5 ± 0.7 Gg. The timeseries reported on fig 1 shows a clear decreasing trend over the entire period, corresponding to annual decline in the bank emission of 4.7% (3 – 6%). From this analysis we can argue that the slow decreasing trend observed by [1] could not be attributable to WEU emissions. The emission distribution, shown in fig 2 a, reveals a hot spot region over Belgium and Netherland.
In contrast with CFC-12 (fig 2 b) where the emissions are spread out over the domain. However, a clear decreasing trend of CFC-12 is reported on timeseries fig 1 b. The average WEU emissions over period 2008 2020 is 1.3 (± 0.35) Gg/yr. Different emission patterns are obtained for the CCl4, where a clear hot spot is located over the south of France. The fig 1 c shows a decreasing trend over the period investigated. A more detailed location is hard to achieve due to the sparsity of the observation network, lack of a priori emission locations, transport model errors, and the low intensity of the emissions estimated. From emission distribution analysis we detected different emissions patterns between these compounds, further investigations are needed to understand the nature of the source regions.

References

[1] Montzka, S. A., Dutton, G. S., Yu, P., Ray, E., Portmann, R. W., Daniel, J. S., Kuijpers, L., Hall, B. D., Mondeel, D., Siso, C., Nance, J. D.,Rigby, M., Manning, A. J., Hu, L., Moore, F., Miller, B. R., and Elkins, J. W.: An unexpected and persistent increase in global emissions of ozone-depleting CFC-11, Nature, 557, 413–417, https://doi.org/10.1038/s41586-018-0106-2, 2018.
[2] Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version6.2, Atmospheric Chemistry and Physics, 5, 2461–2474, https://doi.org/10.5194/acp-5-2461-2005, https://acp.copernicus.org/articles/5/2461/2005/, 2005.
[3] Seibert, P. and Frank, A.: Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode, AtmosphericChemistry and Physics, 4, 51–63, https://doi.org/10.5194/acp-4-51-2004, https://acp.copernicus.org/articles/4/51/2004/, 2004.
[4] Stohl, A., Seibert, P., Arduini, J., Eckhardt, S., Fraser, P., Greally, B. R., Lunder, C., Maione, M., Mühle, J., O'Doherty, S., Prinn, R. G.,Reimann, S., Saito, T., Schmidbauer, N., Simmonds, P. G., Vollmer, M. K., Weiss, R. F., and Yokouchi, Y.: An analytical inversion method for determining regional and global emissions of greenhouse gases: Sensitivity studies and application to halocarbons, AtmosphericChemistry and Physics, 9, 1597–1620, https://doi.org/10.5194/acp-9-1597-2009, https://acp.copernicus.org/articles/9/1597/2009/, 2009.

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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
A manuscript on the West European emissions estimation of main Chlorofluorocarbons (CFCs), trichlorofluoromethane (CFC-11) dichlorodifluoromethane (CFC-12) and Carbon tetrachloride (CCl_4) is currently being prepared
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Future plans

(Please let us know of any imminent plans regarding a continuation of this research activity, in particular if they are linked to another/new Special Project.)

Estimations of European CO_2 and CH_4 emissions fluxes on national scale through top-down approach.
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