LATE REQUEST FOR A SPECIAL PROJECT 2018–2020

MEMBER STATE: Italy

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Project Title: Top-down estimate of chlorofluorocarbon emissions in Europe using a mesoscale inverse modeling technique

If this is a continuation of an existing project, please state the computer project account assigned previously.	**SP** _____	
Starting year: (A project can have a duration of up to 3 years, agreed at the beginning of the project.)	2018	
Would you accept support for 1 year only, if necessary?	YES ✕	NO ☐

Computer resources required for the years: (To make changes to an existing project please submit an amended version of the original form.)		**2018**	**2019**	**2020**
High Performance Computing Facility	(SBU)	1000	600000	600000
Accumulated data storage (total archive volume)[2]	(GB)	5000	20000	25000

Continue overleaf

[1] The Principal Investigator will act as contact person for this Special Project and, in particular, will be asked to register the project, provide an annual progress report of the project's activities, etc.

[2] If e.g. you archive x GB in year one and y GB in year two and don't delete anything you need to request x + y GB for the second project year.

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Extended abstract

The completed form should be submitted/uploaded at https://www.ecmwf.int/en/research/special-projects/special-project-application/special-project-request-submission.

All Special Project requests should provide an abstract/project description including a scientific plan, a justification of the computer resources requested and the technical characteristics of the code to be used.

Requests asking for 1,000,000 SBUs or more should be more detailed (3-5 pages).

Following submission by the relevant Member State the Special Project requests the evaluation will be based on the following criteria: Relevance to ECMWF's objectives, scientific and technical quality, disciplinary relevance, and justification of the resources requested. Previous Special Project reports and the use of ECMWF software and data infrastructure will also be considered in the evaluation process.

All accepted project requests will be published on the ECMWF website.

Introduction

Chlorofluorocarbons (CFCs) are man-made compounds, widely used in the second half of the 20th century, as refrigerants, propellants (in aerosol applications), and solvents. Due to their widespread use and their stability, they have accumulated in the Earth atmosphere, giving rise to important global change phenomena. Indeed, once reached the stratosphere, CFCs release chlorine atoms that catalytically destroy the stratospheric ozone. Moreover, they are radiatively active gases able to absorb the infrared radiation emitted by the Earth surface, thus contributing to the global warming. For this reasons, they were regulated for phase-out under the Montreal Protocol on Substances that Deplete the Ozone Layer and its subsequent amendments. Developed countries were required to begin phasing out CFCs since the end of 1995, while for developing countries the date for phasing out is the end of 2010. In the absence of production, steady declines in CFC emissions are expected as the reservoir of chemicals remaining in existing equipment and products (CFC 'banks') gradually escapes to the atmosphere and diminishes (Montzka et al., 2018). Vollmer et al., 2018 analyzed the rates of CFC-13 (chlorotrifluoromethane), ΣCFC-114 defined as 1,2-dichlorotetrafluoroethane (CFC-114) and 1,1-dichlorotetrafluoroethane (CFC-114a) and CFC-115 (chloropentafluoroethane), measured over the global domain. They show the growth rates of all compounds over the past years significantly larger than would be expected from zero emissions. Moreover Montzka et al., 2018 show that the rate of decline of atmospheric CFC-11 (trichlorofluoromethane) concentrations observed at remote measurement sites was constant from 2002 to 2012, and then slowed by about 50 per cent after 2012. The slower global decline in CFC-11 mole fractions after 2012 represents a perturbation of around 20% in the balance of CFC-11 sources and sinks. The increase in emission of CFC-11 appears unrelated to past production; this suggests unreported new production, which is inconsistent with the Montreal Protocol agreement to phase out global CFC production by 2010. The aim of our project is to estimate the European emissions of CFC-11, CFC-13, CFC-114 and CFC-15, measured at four European measurement sites, from the year 2000 up today: Mt. Cimone CMN (Italy), Jungfraujoch, JFJ (Switzerland) and Mace Head, MHD (Ireland) and Zeppelin ZEP (Ny Ålesund, Spitsbergen). In situ measurements of CFCs together with a Lagrangian dispersion model in conjunction with an analytical inversion method will be applied for this purpose.

Method and application

Measurement stations

In Europe, atmospheric measurement data for a wide range of CFCs are available at four sites: Jungfraujoch (JFJ), Switzerland (CH), Mace Head (MHD), Ireland (IE), Monte Cimone (CMN), Italy (IT), and Zeppelin (ZEP), Norway, Spitzbergen (NO). Through the use of gas chromatographic-mass spectrometric instrumentation (Miller et al., 2008), these stations are providing long term, high frequency and high precision measurements of several halogenated gases. This is the result of a coordination effort that started in 2001 under the European SOGE (System for Observations of Halogenated Greenhouse Gases in Europe) Project. The four European stations are embedded in the AGAGE (Advanced Global Atmospheric Gases Experiment) programme (Prinn et al., 2000), which, together with the NOAA CMDL (National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory)(Montzka et al., 1994) and the NIES (National Institute for Environmental Studies, Japan) (Yokouchi et al., 2006) monitoring networks, represents the most important observation system for a wide range of ozone depleting and climate altering species.

Dispersion model

FLEXible PARTicle-Weather Research and Forecasting FLEXPART is a 3-D Lagrangian particle dispersion model (Stohl et al., 1998; Stohl et al., 2005) (see also http://transport.nilu.no/flexpart) developed to simulate long range and mesoscale dispersion from point sources of hazardous substance, and was validated with several trace experiments (Sthol et al., 1998). Currently FLEXPART is successfully used for case studies of air pollution transport over regional and global domain (Stohl et al., 2009; Stohl et al., 2010; Keller et., 2011; Graziosi et al., 2017).

For our purpose, FLEXPART will be driven with analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF), over the global domain, with a spatial resolution of 1 ° latitude x 1 ° longitude. Due the complexity of topography around the two mountain stations, a nesting wind field of ECMWF (0.125 latitude x 0.125 longitude) will be applied over Alps and north Italy. Comparison between FLEXPART-ECMWF and FLEXPART-WFR (*Weather Research and Forecasting) will be made.* From each measurement stations, tens of thousands of virtual particles will be released and followed back in time back for 20 days to calculate an emission sensitivity, called source-receptor-relationship (SRR) by Seibert and Frank (2004). The SRR concept is important in air quality modelling; it describes the sensitivity of a "receptor" to a "source". The SRR value is proportional to the particle residence time of particles in output grid cells and measures the simulated mixing ratio at the receptor that a source of unit strength in the cell would produce.

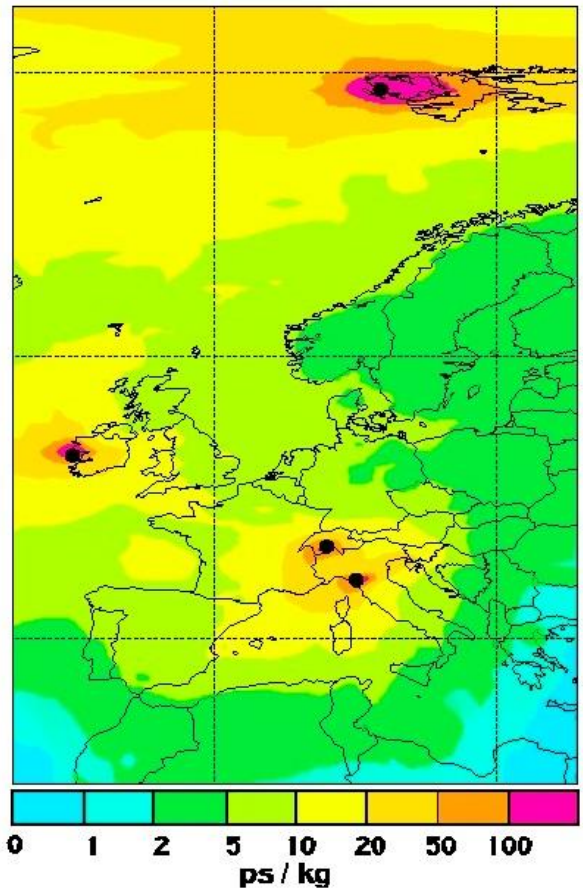

Figure 1. SRR in picoseconds per kilogram (ps kg^{-1}) obtained from FLEXPART 20 d backward calculations averaged over all model calculations over two years (Jan 2008- Dec 2009). Measurement sites are marked with black dots.

Inversion method

In order to identify the CFCs source regions and to give an estimate of the magnitude of such unreported emissions, we will use a Bayesian Inversion method developed by Seibert (2000; 2001) and improved by Eckhardt et al. (Eckhardt et al. 2008) and Stohl et al. (Stohl et al. 2009; 2010).This technique can be applied to determine the spatial-temporal flux distribution of any species for which the atmospheric loss can be described as a linear process. The Bayesian inversion algorithm, determines the model agreement with observations from the measurement sites, taking in to account the model simulations, measurements data, a priori emissions fluxes and the gridded uncertainty on emissions. As no information are available on the localization of European sources, the first part of the analysis will be devoted to the localization itself. The a priori emission field over land area will be set as homogeneously distributed. Furthermore, several tests we could done to improve our analysis, like an increment of a priori uncertainty in correspondence of source localizations on European Pollutant Release and Transfer Register (E-PRTR), investigate the agreement between simulated times series and the observations using different a priori emission fields, test the effect of the station geometry on the inversion results, removing one station at a time, and so on. The estimates provided by this analysis may contribute to constrain the atmospheric budget of CFCs on a regional scale, and improving the understanding of anomalous emissions over the global scale.

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