## SPECIAL PROJECT PROGRESS REPORT

Progress Reports should be 2 to 10 pages in length, depending on importance of the project. All the following mandatory information needs to be provided.

<b>Reporting year</b>	2017			
Project Title:	FLEXPART transport simulations and inverse modelling of atmospheric components			
<b>Computer Project Account:</b>	SPNOFLEX			
Principal Investigator(s):	Espen Sollum			
Affiliation:	NILU- Norwegian Institute for Air Research			
<b>Name of ECMWF scientist(s)</b> <b>collaborating to the project</b> (if applicable)	Sabine Eckhardt, Massimo Cassiani, Rona Thompson, Thomas Hamburger, Henrik Grythe, Ignacio Pisso, Arve Kylling, Andreas Stohl, Christine Groot Zwaaftink, Nikolaos Evangeliou, Espen Sollum			
Start date of the project:	2017			
Expected end date:	2019			

# **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)	50000	60300	100000	23528
Data storage capacity	(Gbytes)	150	150	150	150

## Summary of project objectives

The Lagrangian particle dispersion model FLEXPART is run on ECMWF data to explore the transport and dispersion of various atmospheric constituents from greenhouse gases, aerosols like black carbon to volcanic ash released during eruptions. The model is used with various inversion techniques to infer emission estimates of many atmospheric compounds. This helps improving transport simulations of these substances and to understand their contribution and effects on the climate system.

## Summary of problems encountered (if any)

**Summary of results of the current year** (from July of previous year to June of current year)

This section should comprise 1 to 8 pages and can be replaced by a short summary plus an existing scientific report on the project

The following 4 summaries highlights our use of ECMWF data during the previous year:

- 1. Black Carbon (BC) in Arctic surface and snow
- 2. Determination of <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I emissions using inverse modelling and an updated measurement database
- 3. Modelling mineral dust in the Arctic and Iceland
- 4. Quantifying the mass loading of particles in an ash cloud remobilized from tephra deposits on Iceland

#### 1) Black Carbon (BC) in Arctic surface and snow

#### N. Evangeliou, A. Stohl

The concentrations of BC were simulated with version 10 of the LPDM FLEXPART (FLEXible PARTicle dispersion model) (Stohl et al., 2005). The model was driven with operational meteorological analyses every three hours from the European Centre for Medium-Range Weather Forecasts (ECMWF, operational fields). Computational particles released from the measurement locations were tracked back in time in FLEXPART's "retroplume" mode (Stohl et al., 2003). Simulations extended over 30 d back in time, sufficient to include most aerosol emissions arriving at the station, given a typical BC lifetime (~1 week). This enabled identifying where the measured BC came from and allowed quantification of BC source contributions. FLEXPART simulations were performed every hour during the cruise, with particles released from small boxes covering the latitude and longitude ranges of the ship track during the hour (Fig. 1). The FLEXPART retroplumes consist of an emission sensitivity (often also called source-receptor relationship), which yields a simulated concentration in the receptor box when multiplied with gridded emissions from an inventory.



**Fig. 1.** (a) Time series of equivalent black carbon (EBC) during the expedition cruise. (b) Age spectra of modeled BC (colors) showing the contribution of emissions each day back in time to the surface concentration of BC. Hourly means of measured BC concentrations are shown as a black line. (c) Contribution from different emission source types to the BC surface concentrations (BB: biomass burning, WST: waste burning, IND: industrial combustion, TRA: transportation, ENE: power plants, energy conversion, and extraction, DOM: residential and commercial combustion from GFEDv3.1 and ECLIPSE inventories). More details in Popovicheva *et al.* (2017).

The simulations for the snow BC (Fig. 2) were conducted using a new feature of FLEXPART that reconstructs wet and dry deposition with backward simulations. This new feature is an extension of the aforementioned retroplume mode. To reconstruct wet deposition amounts of BC, computational particles were released at altitudes of 0 to 20 km at the locations where snow samples were taken, whereas to reconstruct dry deposition, particles were released between the surface and 30 m at these locations. All released particles together represent a unity deposition amount, which was converted immediately (i.e., upon release of a particle) to atmospheric concentrations using the deposition intensity as characterized either by dry deposition velocity or scavenging rate. The concentrations were subsequently treated as in normal retroplume backward tracking to establish emission sensitivity between the emissions and deposited amounts. The ending time of the particle release was the time at which the snow sample was collected, whereas the beginning time was set as the time when the ECMWF precipitation at the sampling site, accumulated backward in time, was equal to the water equivalent of the snow sample, up to the specified sampling depth.

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**Fig. 2.** Contribution from the various emission categories considered in the ECLIPSE and GFED inventories (BB: biomass burning, WST: waste burning, IND: industrial combustion, TRA: surface transportation, ENE: emissions from energy conversion, DOM: residential and commercial combustion, FLR: gas flaring) to simulated BC concentrations in snow during the 2014, 2015 and 2016 in Western Siberia and northwestern European Russia. Bars show the relative source contribution (0 –100%, right axis) and are sorted, from left to right, from the northernmost to the southernmost measurement location (coordinates are reported on the bottom as longitude/latitude). Measured EC concentrations in snow are reported with open circles, whereas modelled BC is shown with open rectangles (left axis). More details in Evangeliou *et al.* (2017).

## 2) Determination of <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I emissions using inverse modelling and an updated measurement database

N. Evangeliou, T. Hamburger, A. Stohl

We have used Bayesian inversion to determine of the source term of the radionuclides <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I released after the Chernobyl accident. The accident occurred on 26 April 1986 in the Former Soviet Union and released about 10<sup>19</sup> Bq of radioactive materials that were transported as far away as the USA and Japan. Thereafter, several attempts to assess the real magnitude of the emissions were made that were based on the knowledge of the core inventory and the levels of the spent fuel. More recently, when modelling tools were further developed, inverse modelling techniques were applied to the Chernobyl case for source term quantification. However, high quality measurements, that are essential for inverse modelling, were not made available except for a few sparse activity concentration measurements far from the source and far from the main direction of the radioactive fallout. For the first time, we apply Bayesian inversion of the Chernobyl source term using not only activity concentrations, but also deposition measurements from the most recent public dataset (Evangeliou et al., 2016).



**Fig. 3.** Posterior emissions of <sup>134</sup>Cs, <sup>137</sup>Cs and <sup>131</sup>I against uncertainty plotted daily, as well as per time–step (3 h) from 26 April to 7 May 1986. On the right axis the vertical distribution of the emissions at altitudes 0-1 km (yellow), 1-2 km (beige) and 2-3 km (turquoise) is plotted as shaded background colours.

As regards to our inverse modelling results (Fig. 3), emissions of <sup>134</sup>Cs were estimated to be 80 PBq or 30–50 % higher than what was previously published. From the released amount of <sup>134</sup>Cs, about 70 PBq were deposited all over Europe. Similar to <sup>134</sup>Cs, emissions of 137Cs were estimated as 86 PBq, in the same order with previously reported results. Finally, <sup>131</sup>I emissions of 1365 PBq were found, which are about 10 % less than the prior total releases. The inversion pushes the injection heights of the three radionuclides to higher altitudes (up to about 3 km) than previously assumed ( $\approx 2.2$  km) in order to better match both concentration and deposition observations over Europe. The results were of the present inversion were confirmed using an independent Eulerial model, for which deposition patterns were also improved when using the estimated posterior releases. Although the independent model tends to underestimate deposition in countries that are not in the main direction of the plume, it reproduces country levels of deposition very efficiently. The results were also tested for robustness against different set-ups of the inversion through sensitivity runs. More information can be found in Evangeliou et al. (2017a).

#### References

Evangeliou, N., Hamburger, T., Talerko, N., Zibtsev, S., Bondar, Y., Stohl, A., Balkanski, Y., Mousseau, T. A. and

Møller, A. P.: Reconstructing the Chernobyl Nuclear Power Plant (CNPP) accident 30 years after. A unique database of air concentration and deposition measurements over Europe, Environ. Pollut., (August),

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Evangeliou, N., Hamburger, T., Cozic, A., Balkanski, Y. and Stohl, A.: Inverse modelling of the Chernobyl source term using atmospheric concentration and deposition measurements, Atmos. Chem. Phys. Discuss., (April), 1–43, doi:10.5194/acp-2017-330, 2017a.

Evangeliou, N., Shevchenko, V. P., Yttri, K. E., Sollum, E., Pokrovsky, O. S., Kobelev, V. O., Vladimir, B., Lobanov, A. A., Starodymova, D. P., Vorobiev, S. N., Thompson, R. L., Stohl, A., Toulouse, G. E. and Belin, E.: Origin of elemental carbon in snow from Western Siberia and northwestern European Russia during winter – spring, Atmos. Chem. Phys. Discuss., 1–33, doi:10.5194/acp-2017-542, 2017b.

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Stohl, A., Forster, C., Eckhardt, S., Spichtinger, N., Huntrieser, H., Heland, J., Schlager, H., Wilhelm, S., Arnold, F. and Cooper, O.: A backward modeling study of intercontinental pollution transport using aircraft measurements, J. Geophys. Res. Atmos., 108(D12), 4370, doi:10.1029/2002JD002862, 2003.

Stohl, A., Forster, C., Frank, A., Seibert, P. and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5(9), 2461–2474, doi:10.5194/acp-5-2461-2005, 2005.

### 3) Modelling mineral dust in the Arctic and Iceland

#### C. Groot Zwaaftink, H. Grythe, S. Eckhardt, A. Stohl

Mobilization, atmospheric transport and deposition of mineral dust in the Arctic have been studied with FLEXPART. A module to estimate dust emission from source regions, called FLEXDUST, was developed. In this module we use meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) to estimate mineral dust availability and emission. In a series of global simulations for the years 2010-2012 we analysed the seasonal variation of dust emission. Simulations showed that emission from Arctic source regions peaks in late summer/fall, likely due to limited snow cover. Atmospheric transport was calculated with FLEXPART, driven by ECMWF analysis fields. Seasonal variation of atmospheric dust load in the Arctic was strong and peak dust loads were found in spring. Dust loads were dominated by dust from remote source regions. Near the surface, however, simulations showed that local sources contribute substantially to dust concentrations and therefore dust deposition in the Arctic. Dust deposition patterns (Figure 1), reveal that transport of dust from northern high-latitude sources is limited and large fractions are deposited close to the source regions.



Figure Simulated contribution of a selection of source regions to mean deposition of mineral dust in the Arctic in years 2010-2012 (Figure from Groot Zwaaftink et al., 2016).

Following this, we studied the high-latitude dust sources in Iceland in more detail. Here, we run FLEXDUST and FLEXPART simulations for a single year at high resolution and studied interannual variability over 27 years at a coarser resolution. For 2012, hourly ECMWF analysis data at 0.2°-resolution were used in FLEXPART simulations. The model is able to predict the timing of dust events consistent with particulate matter observations at short and long distances to the dust sources. Some inconsistencies were found for small dust sources, related to source depletion and snow cover representation. For the period 1990-2016, FLEXPART was driven with ERA Interim Reanalysis fields. These long-term simulations showed that annual dust emission from Iceland is on average about 4.3 Tg. Considering the small size of dust sources (<20.000 km<sup>2</sup>) this is a large amount and Icelandic dust sources thus appear to be as active as parts of the Sahara. Most of the dust mobilized in Iceland is deposited in the ocean, both north-east and south of Iceland, while some dust also reaches the Greenland Ice sheet (Figure 2). Further study showed that dust deposition on glaciers in Iceland strongly affects glacier melt rates (Wittmann et al., 2017).



Figure Simulated annual mean deposition of dust originating from Iceland in years 1990-2016 based on FLEXPART simulations (figure from Groot Zwaaftink et al., 2017).

#### 4) Quantifying the mass loading of particles in an ash cloud remobilized from tephra deposits on Iceland

Frances Beckett , Arve Kylling , Guðmunda Sigurðardóttir, Sibylle von Löwis , and Claire Witham

On 16–17 September 2013 strong surface winds over tephra deposits in southern Iceland led to the resuspension and subsequent advection of significant quantities of volcanic ash. The resulting resuspended ash cloud was transported to the south-east over the North Atlantic Ocean and, due to clear skies at the time, was exceptionally well observed in satellite imagery. We use satellite-based measurements in combination with radiative transfer and dispersion modelling to quantify the total mass of ash resuspended during this event. Typically ash clouds from explosive eruptions are identified in satellite measurements from a negative brightness temperature difference (BTD) signal; however this technique assumes that the ash resides at high levels in the atmosphere. Due to a temperature inversion in the troposphere over southern Iceland during 16 September 2013, the resuspended ash cloud was constrained to altitudes of < 2 km a.s.l. We show that a positive BTD signal can instead be used to identify ash-containing pixels from satellite measurements. The brightness temperature (BT) varies with the amount of water vapour in the atmosphere, the atmospheric temperature profile, and the temperature of the underlying surface. For these parameters analysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) were utilized to estimate water vapour effects on the brightness temperature difference signal used to identify ash affected pixels. Examples of BTDs for the remobilization event is shown in Fig. 1. The timing and location of the ash cloud identified using this technique from measurements made by the Visible Infrared Imaging Radiometer Suite (VIIRS) on board the Suomi National Polarorbiting Partnership (NPP) satellite agree well with model predictions using the dispersion model NAME (Numerical Atmospheric-dispersion Modelling Environment). Total column mass loadings are determined from the VIIRS data using an optimal estimation technique which accounts for the low altitude of the resuspended ash cloud and are used to calibrate the emission rate in the resuspended ash scheme in NAME. The column mass loading retrieval used look-uptables calculated using area-averaged ECMWF water vapour, pressure and temperature profiles. Considering the tephra deposits from the recent eruptions of Eyjafjallajökull and Grímsvötn as the potential source area for resuspension for this event, we estimate that  $\sim 0.2$  Tg of ash was remobilized during 16–17 September 2013.



VIIRS 16/09/2013 05:24 UTC

VIIRS 16/09/2013 12:00 UTC

VIIRS 16/09/2013 03:42 UTC

Figure 1. VIIRS BTDs for pixels identified as resuspended ash, 16-17 September 2013.

VIIRS 16/09/2013 02:06 UTC

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

- Popovitseva, O. B., Evangeliou, N., Eleftheriadis, K., Kalogridis, A. C., Sitnikov, N., Eckhardt, S., and Stohl A.: Black Carbon Sources Constrained by Observations in the Russian High Arctic, Environ. Sci. Technol., 51 (7), 3871–3879, doi:10.1021/acs.est.6b05832, 2017.
- Xu, J., Martin, R. V., Morrow, A., Sharma, S., Huang, L., Leaitch, W. R., Burkart, J., Schulz, H., Zanatta, M., Willis, M. D., Henze, D. K., Lee, C. J., Herber, A. B., and Abbatt, J. P. D.: Source attribution of Arctic black carbon constrained by aircraft and surface measurements, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-236, 2017.
- Evangeliou, N., Hamburger, T., Cozic, A., Balkanski, Y., and Stohl, A.: Inverse modelling of the Chernobyl source term using atmospheric concentration and deposition measurements, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-330, 2017.
- Evangeliou, N., Shevchenko, V. P., Yttri, K. E., Eckhardt, S., Sollum, E., Pokrovsky, O. S., Kobelev, V. O., Korobov, V. B., Lobanov, A. A., Starodymova, D. P., Vorobiev, S. N., Thompson, R. L., and Stohl, A.: Origin of elemental carbon in snow from Western Siberia and northwestern European Russia during winter–spring 2014, 2015 and 2016, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-542, 2017.
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- Groot Zwaaftink, C.D. Ó. Arnalds, P. Dagsson-Waldhauserova, S. Eckhardt, J. M. Prospero, A. Stohl (2017), Temporal and spatial variability of Icelandic dust emission and atmospheric transport, under review for *Atmospheric Chemistry and Physics*. DOI:10.5194/acp-2017-290
- Wittmann, M., C. D. Groot Zwaaftink, L. Steffensen Schmidt, S. Guðmundsson, F. Pálsson, O. Arnalds, H. Björnsson, T. Thorsteinsson, and A. Stohl (2017), Impact of dust deposition on the albedo of Vatnajökull ice cap, Iceland, *The Cryosphere*, 11(2), 741-754. DOI:10.5194/tc-11-741-2017
- Groot Zwaaftink, C. D., H. Grythe, H. Skov and A. Stohl (2016), Substantial contribution of northern high-latitude sources to mineral dust in the Arctic, *Journal of Geophysical Research*, 121, DOI: 10.1002/2016JD025482
- Beckett, F., Kylling, A., Sigurðardóttir, G., von Löwis, S. and Witham, C., 'Quantifying the mass loading of particles in an ash cloud remobilized from tephra deposits on Iceland', Atmospheric Chemistry and Physics, 7, 17, 4401-4418, <u>http://www.atmos-chemphys.net/17/4401/2017/</u>, doi:10.5194/acp-17-4401-2017, 2017.

## Summary of plans for the continuation of the project

(10 lines max)

ECMWF data will be continued to be used within the various inversion frameworks for estimating greenhouse gas emissions, radionuclide emissions and volcanic emissions, and subsequent FLEXPART transport simulations using the inverted sources.