

*Contribution; Version for proceedings of ETH 2014 conference: 25 July 14; 10:00*

**Fukushima Xe-133 detection at the tropopause: Implications for very efficient no-particle formation at the tropopause from lifted SO<sub>2</sub> of East-Asian origin.**

H. Schlager (1), **F.Arnold (1,2)\***, H.Aufmhoff (1), R.Baumann (1), H. Simgen (2), S.Lindemann (2), L. Rauch (2), F. Kaether (2), C.Schlosser (3), and U. Schumann (1)

**\* Speaker**

(1) Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, Germany

(2) Max-Planck-Institut für Kernphysik, Heidelberg, Germany

(3) Bundesamt für Strahlenschutz, Freiburg, Germany

**Abstract**

We report determined airborne measurements, at the tropopause, of the radioactive noble gas Xe-133, accidentally released by the major nuclear power plant disaster at Fukushima (mostly in the period 11-15 March 2011). Our unique measurements, representing the first and so far only airborne Xe-133 measurements, were possible by employing the most sensitive Xe-133 detection technique, developed at one of our laboratories (MPIK-Heidelberg). The airborne measurements allowed us to use Xe-133 as an ideal tracer for WCB induced transport of ground-level air to the tropopause, followed by intercontinental high-altitude transport. Our measurement and our accompanying air transport model simulations reveal that parts of the Fukushima Xe-133 plume were lifted by the North-East-Pacific WCB (Warm Conveyor Belt) flow of a cyclone. On 16 March, large parts of the Xe-133 plume experienced WCB induced rapid lift (within about 12 hours) to about 10 km altitude. Hereafter, the plume was carried by high-altitude winds across the North-Pacific, North-America, and the North-Atlantic. During this intercontinental transport, the plume experienced additional moderate ascent. Already on 20 March the plume reached the Upper troposphere and lowest stratosphere over Central-Europe, where our first research aircraft flight took place on 23 March 2011. Our second flight took place on 14 April when the, meanwhile considerably diluted, plume had already spread over large

the parts of the North-Hemisphere. Our investigations prove unambiguously that North-West-P

## Scientific background

The tropopause (TP) region, and particularly the TIL (Tropopause Inversion Layer), located just above the TP) represents an interesting atmospheric region, offering ideal conditions for SO<sub>2</sub> conversion to the key aerosol precursor gas-phase sulfuric acid and resulting sulfate aerosol formation and growth (c.f. [Schlager et al, 2014](#); [Arnold et al, 2014](#)). However, the SO<sub>2</sub> input into the TIL is not well known. This is particularly true for SO<sub>2</sub> input via anthropogenic SO<sub>2</sub> lift from ground-level SO<sub>2</sub> sources (mostly fossil fuel combustion).

The TIL is present in middle latitudes and extends from the thermal tropopause (TTP) to a maximum of about 2 km above the TTP. The TIL thickness varies with season and latitude. Important details of TIL formation are as yet not fully understood.

Potential sources of SO<sub>2</sub> present in the TIL are: airplanes cruising in or in the vicinity of the TIL, whose engines release SO<sub>2</sub> as a combustion product of fuel sulfur; volcanic SO<sub>2</sub> input; lift of SO<sub>2</sub> from ground-level SO<sub>2</sub> sources.

Lift of SO<sub>2</sub> from ground-level SO<sub>2</sub> sources to the TIL is not well known. The efficiency of vertical air transport from the planetary boundary layer of major anthropogenic SO<sub>2</sub> emission regions, which are located mostly in middle latitudes of the northern hemisphere, is only poorly known. Also poorly known is heterogeneous SO<sub>2</sub> loss by cloud processes, occurring during lift. Relevant heterogeneous SO<sub>2</sub> loss processes include washout, rainout, and liquid-phase conversion to sulfate.

The strongest ground-level fossil fuel combustion related SO<sub>2</sub> releases to the atmosphere are presently occurring in middle-latitude East-Asia (East-China/ Korea/ Japan; hereafter: CKJ region), followed by Europe and eastern North-America (c.f. [EDGAR archive](#); [new Paper](#)). Efficient transport from the JKC region ground level SO<sub>2</sub>-sources to the tropopause and the TIL may be induced by warm conveyor belts (WCBs), which are particularly pronounced over the middle latitude West-Pacific. WCB activity has a pronounced seasonal variation. In the northern hemisphere, WCB activity is strongest in the period November-April (c.f. [Eckhardt et al, Madonna et al, 2014](#)).

WCBs are usually associated with middle latitude cyclones. Air mass transport model simulations suggest that a WCB may lift, within about about 12-24 hours, planetary boundary lay

er air to the tropopause.

Already previously, we have observed SO<sub>2</sub>-rich pollution plumes in the middle and upper troposphere, which according to our previous model simulations, had been lifted by East-Asian WCBs. However, so far no such measurements have been reported for the tropopause region and the TIL. The first airborne mass spectrometric SO<sub>2</sub> measurements in the entire TIL-altitude range (made by DLR aboard HALO; see above) will be published in a separate paper. The present paper, reports unique measurements at the TP and in the TIL, of Fukushima Xe-133, representing an ideal tracer for North-West-Pacific WCB mediated transport.

### **Accidental Xe-133 release at Fukushima**

The nuclear disaster of March 2011 resulted in the release to the atmosphere of numerous radionuclides. Among these, was also the radioactive noble gas Xe-133, which was released preferably during the period 11-15 March, from nuclear reactor blocks of the Fukushima/Daiichi nuclear power plant complex. Fukushima Xe-133 represents an ideal atmospheric transport tracer since it is chemically inert and therefore represents a tracer, which is not affected by chemical processes and cloud processes. Its e-folding lifetime against radioactive decay (7.5 days; half-life: 5.25 days) is precisely known and is approximately comparable to the time required for WCB mediated transport in the UTLS (Upper Troposphere/Lower Stratosphere) from Fukushima to Europe. Importantly, the Xe-133 lifetime, at and above the tropopause, is also approximately comparable to the time scale for OH-induced SO<sub>2</sub> conversion in the (e-folding lifetime of SO<sub>2</sub>: about 5-10 days). Hence, the concentration ratio lifted SO<sub>2</sub>/Xe-133 should remain approximately constant during quasi-horizontal transport in the TIL.

Nuclear power plants, using the nuclear fuel U-235, generate large amounts of Xe-133 by neutron induced U-235 fission. In normal operational conditions, Xe-133 remains inside the fuel rods and/or pressure vessel of the reactor, where it undergoes relatively rapid radioactive decay. During a nuclear power plant malfunction, with loss of cooling of the solid nuclear fuel rods, the fuel rod temperature rapidly increases and as a consequence Xe-133 is readily released from the fuel rods. Interaction of water with the hot fuel rods may lead to catalytic H<sub>2</sub> generation, which may lead to an H<sub>2</sub>/O<sub>2</sub> explosion. Such an explosion may damage the pressure vessel, resulting in uncontrolled Xe-133 release to the atmosphere. In order to avoid an H<sub>2</sub>/O<sub>2</sub> explosion, the pressure vessel may occasionally be vented in a controlled way, leading to rapid release of H<sub>2</sub> and Xe-133 to the atmosphere.

Release of Xe-133 at the Fukushima/Daiichi (FD) nuclear power plant complex (from four reactor blocks) took place mostly in the period 11 to 15 March 2011 (Stohl et al., 2011), when several H<sub>2</sub>/O<sub>2</sub> explosions and determined reactor pressure vessel venting operations occurred. The total initial inventory of Xe-133 present in the 4 nuclear reactor blocks is estimated to be about 1.2-1.6 E(19) Bq. The reconstructed history of Xe-133 release at Fukushima, as previously been reported (c.f. Stohl et al., 2011).

### **Investigation-Strategy**

Since the Xe-133 release period (mostly 11-15 March) was still within the Northern-Hemisphere WCB high-season (November-April), there was a realistic chance that parts of the Fukushima Xe-133 plume may become lifted to the UTLS (Upper Troposphere Lower Stratosphere) by a West-Pacific WCB. Due to fast westerly-winds, prevailing in the UTLS, the lifted plume would be advected to the European UTLS within only a few days. Therefore, we recognized the unique opportunity of using FD Xe-133 for an ideal tracer experiment, aiming for an investigation of East-Asian WCB mediated anthropogenic pollutants transport.

Already one day after the first Fukushima nuclear power plant explosion had been reported by the media (on 11 March 2010), we responded quickly by starting predictive model simulations of the transport and dispersion of the Fukushima Xe-133 plume. A particular objective of the predictive model simulations was to find out whether the Fukushima radioactive plume experienced WCB induced lift to the UTLS and particularly to the TIL. Furthermore, we started activities aiming for airborne measurements of Fukushima Xe-133 using the German research aircraft FALCON (operated by DLR).

### **Summary and conclusions**

The major findings of our investigation may be summarized as follows:

- 1) Unique airborne detection of the Fukushima radionuclide Xe-133 at the tropopause over Central-Europe.
- 2) Our accompanying model simulations of the Fukushima Xe-133 plume compare are similar to our measured Xe-133.
- 3) Our air mass model simulations of the back-trajectories of air masses probed at the tropopause over Europe do not exactly lead to Fukushima, but at least lead to a region 20

0 km east of Fukushima.

From our experimental and modeling investigations we draw the following major conclusions :

- a) Parts of the Fukushima Xe-133 plume were rapidly (within about 12 hours on 16 March 2011) lifted to the tropopause. Hereafter, the lifted plume experienced intercontinental transport to Europe, where it arrived on 20 March 2011.
- b) During intercontinental transport, some additional ascent took place, even to the TTL.
- c) The initial large-scale lift was, without doubt, induced by a West-Pacific WCB (Warm conveyor Belt) associated with a pronounced cyclone.
- d) Radioactive radiation doses received by passengers and crewmembers of airliners, penetrating the Fukushima plume at typical cruise altitudes, are small compared to the radiation doses aviation passengers receive from galactic cosmic radiation.
- e) The most important conclusion from our investigation is: We have found an efficient transport path connecting the region with the strongest WCBs with the region of the largest fossil fuel combustion related pollution sources. This is particularly true for the fossil fuel combustion generated gas SO<sub>2</sub>, which represents an important precursor of nanoparticles and grown aerosol particles.

#### **References:**

**See full paper Schlager et al, submitted 2014 (for Information contact Prof. F. Arnold (frank.arnold@mpi-hd.mpg.de))**

#### **Acknowledgements:**

We acknowledge the scientific and technical support by several institutions, including DLR-IPA, DLR-Flugbetrieb, MPIK-Heidelberg, and Bundesamt für Strahlenschutz. Part of the proj

ect was funded by DLR, MPG, Bfs, and BMBF. One of us (F.A.) acknowledges partial funding via a Max-Planck research award in Physics.



# Figure 1

Title

Authors

Institutions



# ***Fukushima Xe-133 detection at the tropopause: Implications for very efficient nano-particle formation at the tropopause from lifted SO<sub>2</sub> of East-Asian origin***

H.Schlager (1), **F.Arnold** \*(2,1), R.Baumann (1), H.Aufmhoff (1), A.Reiter (1),  
H.Simgen (2), L.Rauch (1), S. Lindemann (2), F.Käther (2), and U. Schumann (1)  
(\* Speaker)

(1) DLR Oberpfaffenhofen, Germany

(2) Max-Planck-Institut für Kernphysik, Heidelberg, Germany



Oral presentation at 18th ETH Conference on „Combustion Generated Nanoparticles“  
Zuerich (Switzerland), 23 June 2014

# Figure 2

*Research air planes used:*

- FALCON (operated by DLR)
- HALO (operated by DLR)

**FALCON**



**HALO**

# Figures 3a-h

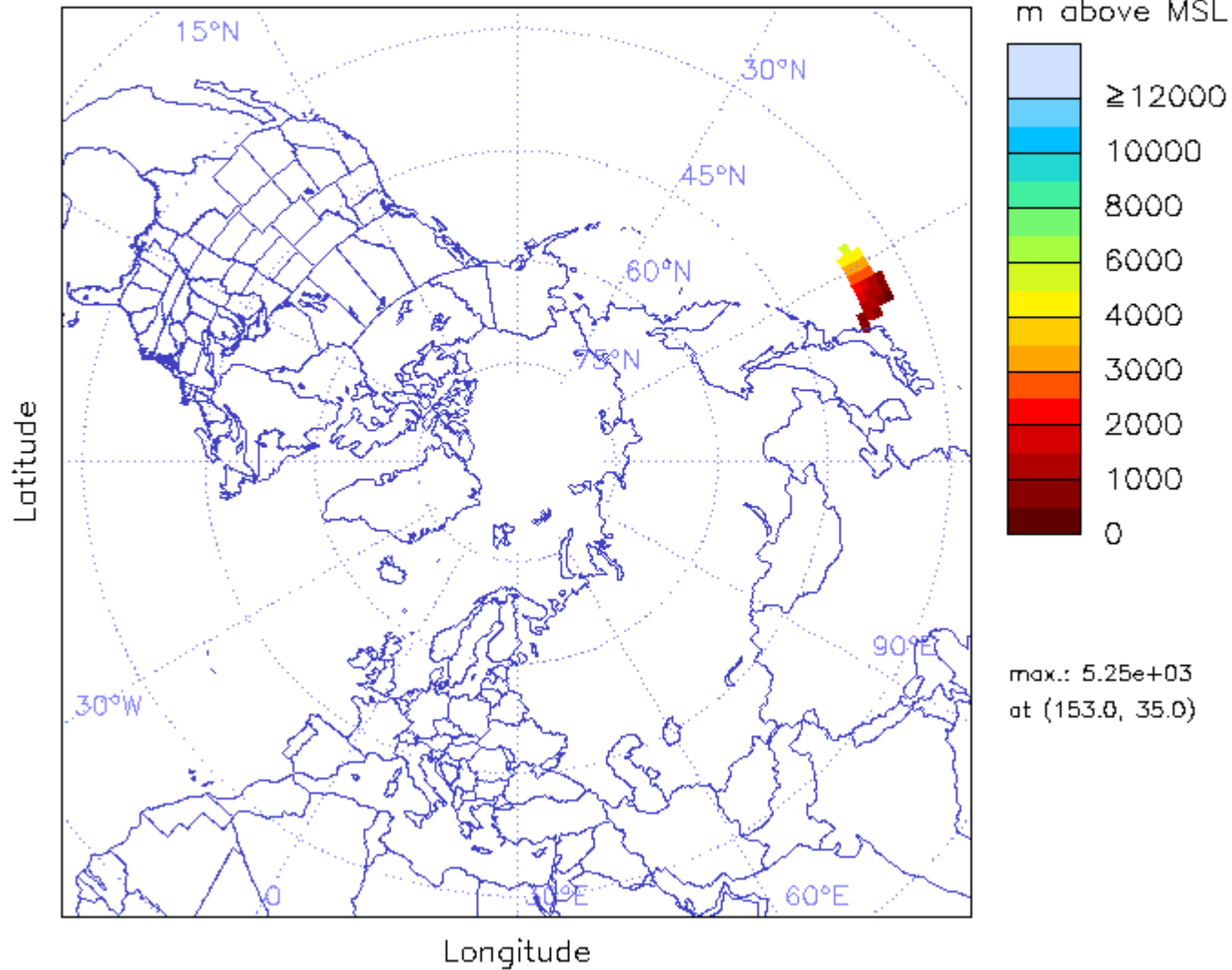
- Model-1 simulation of the evolution of the Fukushima Xe-133 plume. Plotted is a time-series of snapshots of the median-height of the plume for the period 12 March 2011 – 14 April 2011

NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 12 Mar

median height of distribution, 00:00 UTC 12 Mar

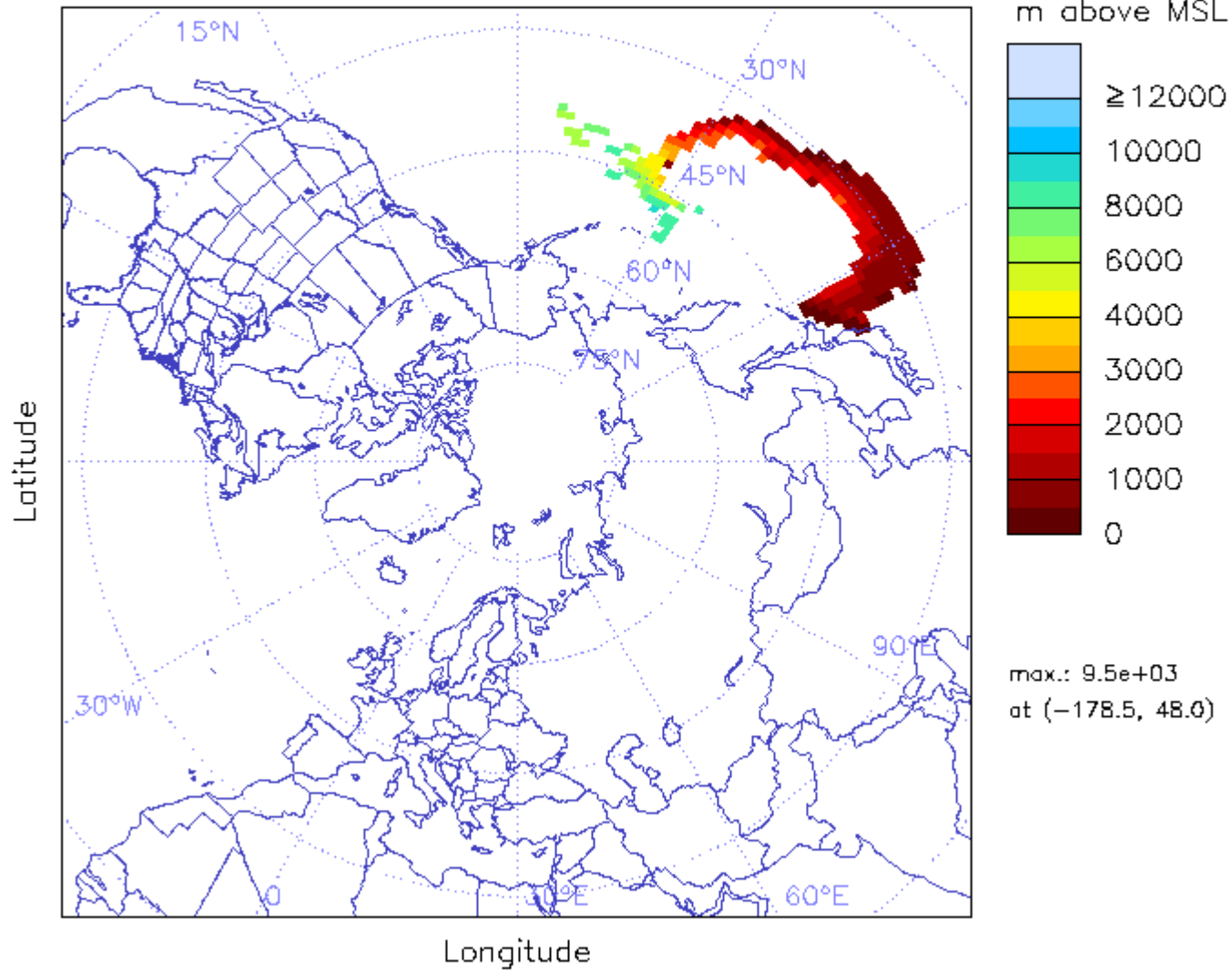


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 13 Mar

median height of distribution, 00:00 UTC 13 Mar

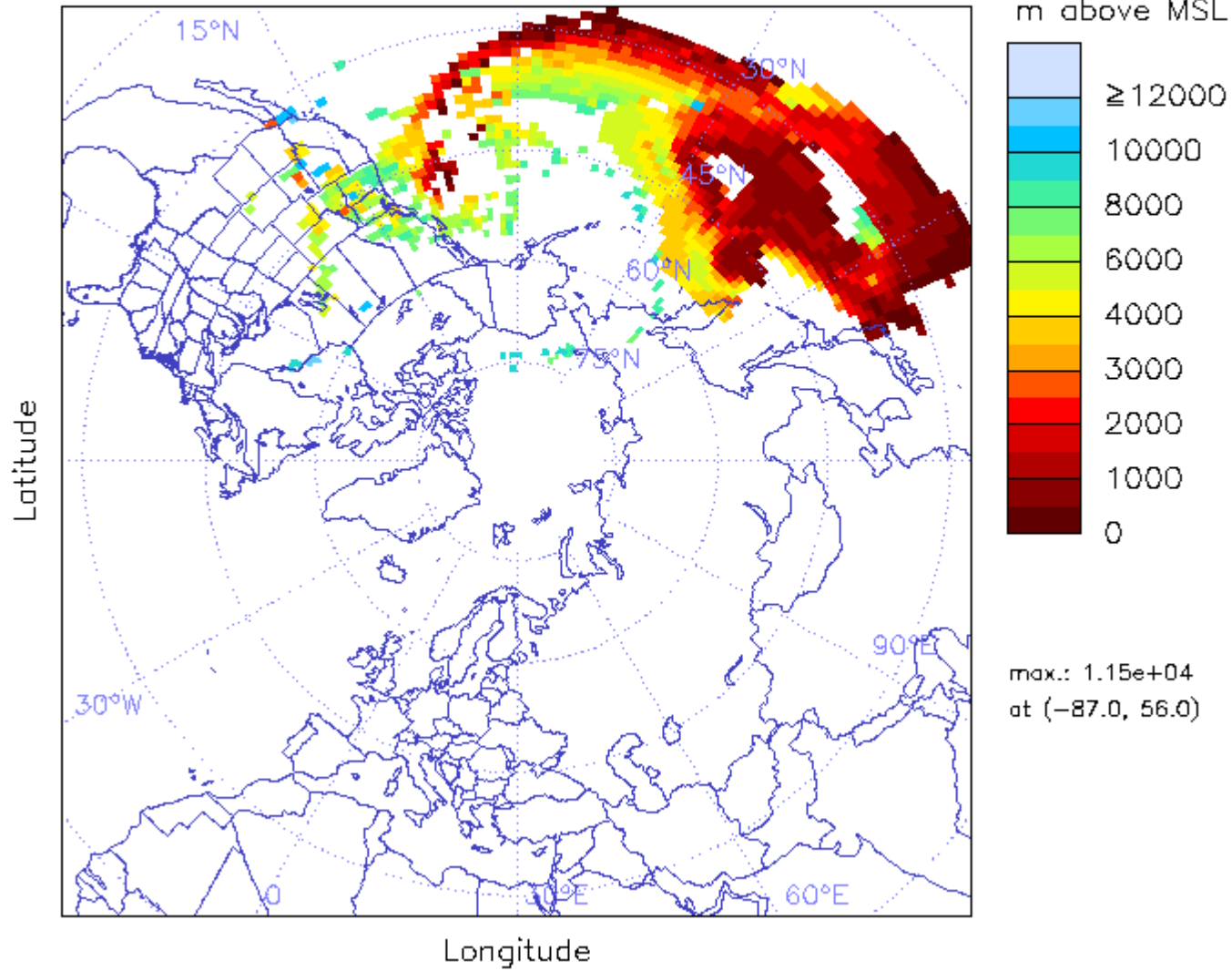


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 15 Mar

median height of distribution, 00:00 UTC 15 Mar

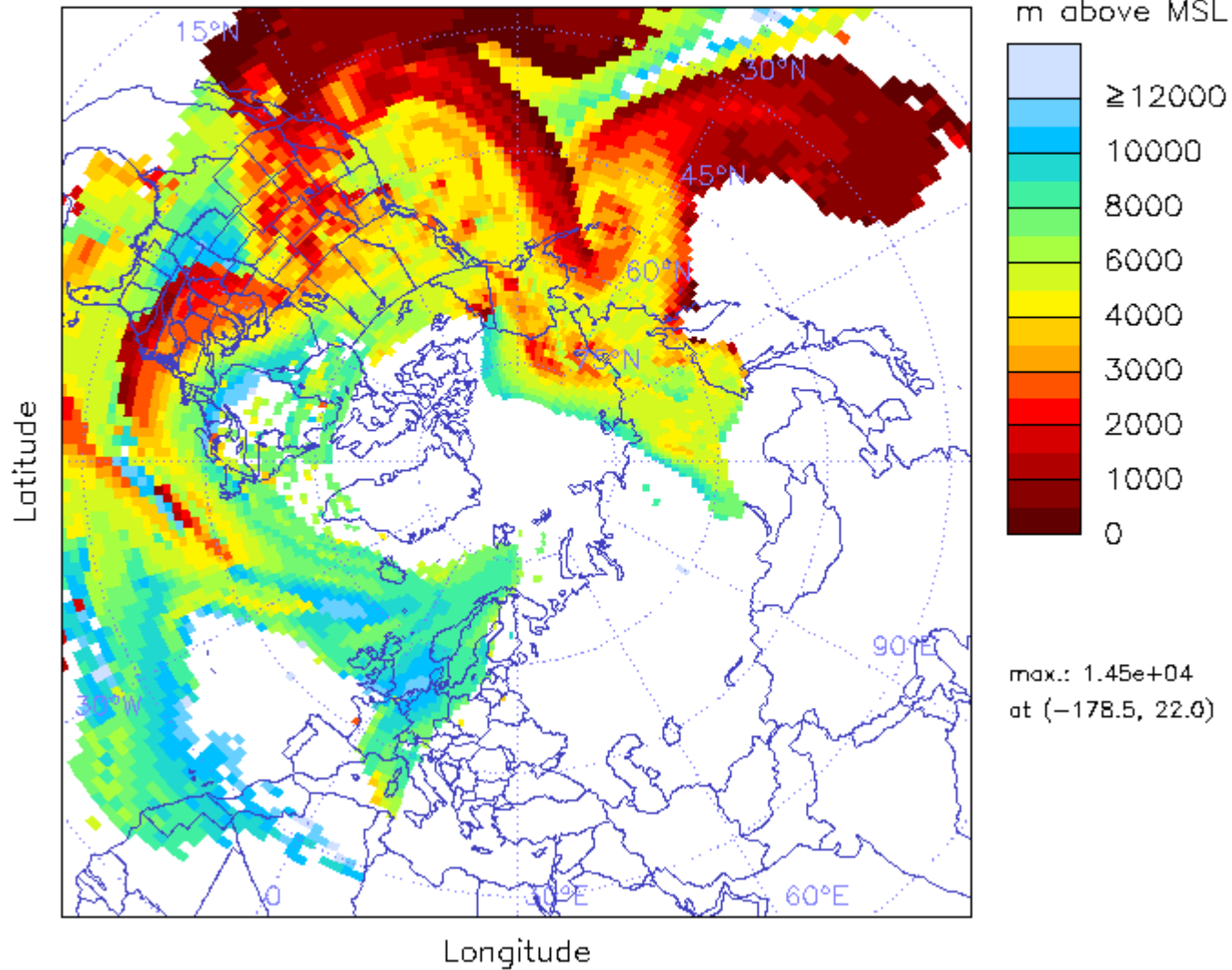


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 20 Mar

median height of distribution, 00:00 UTC 20 Mar



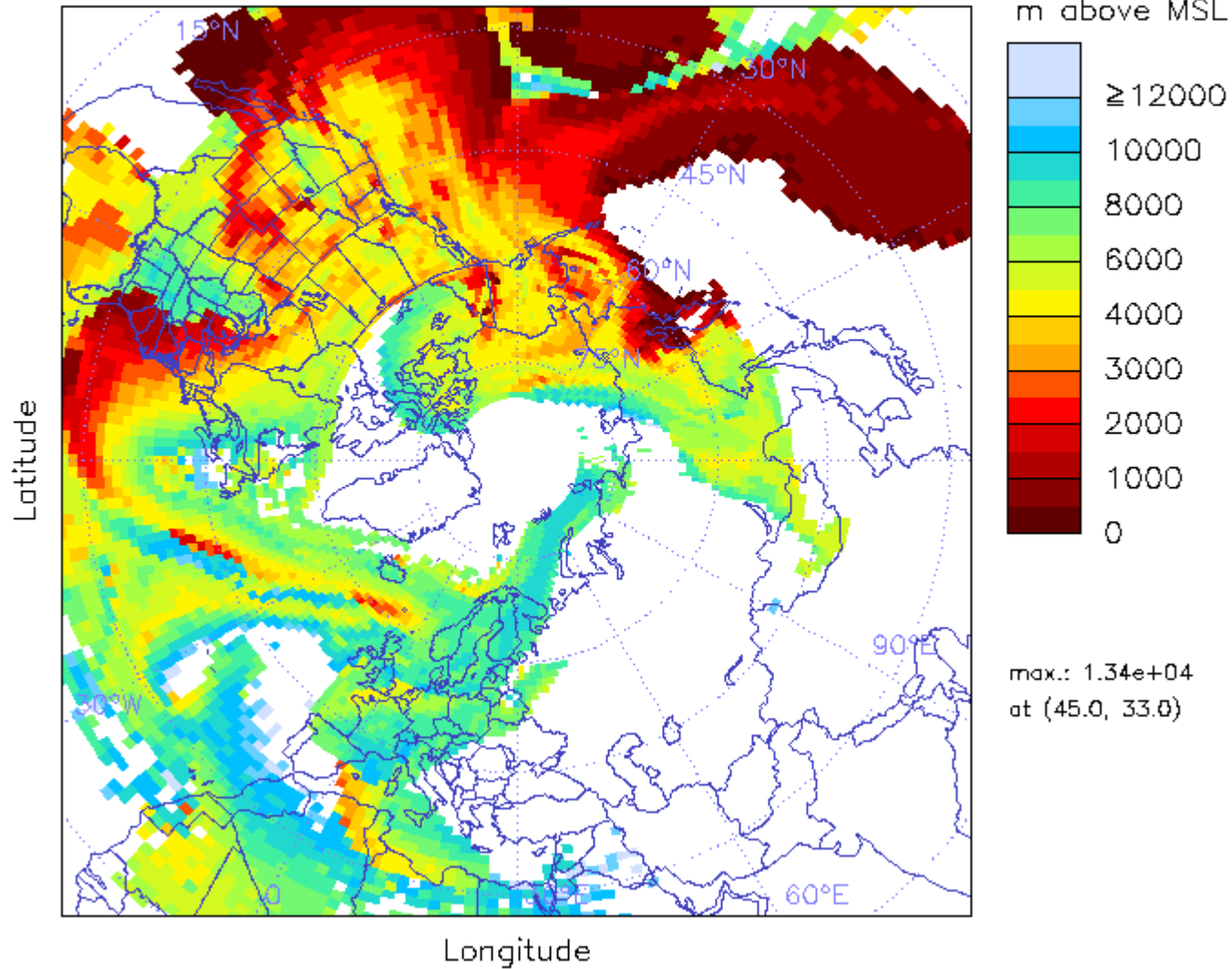


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 21 Mar

median height of distribution, 00:00 UTC 21 Mar

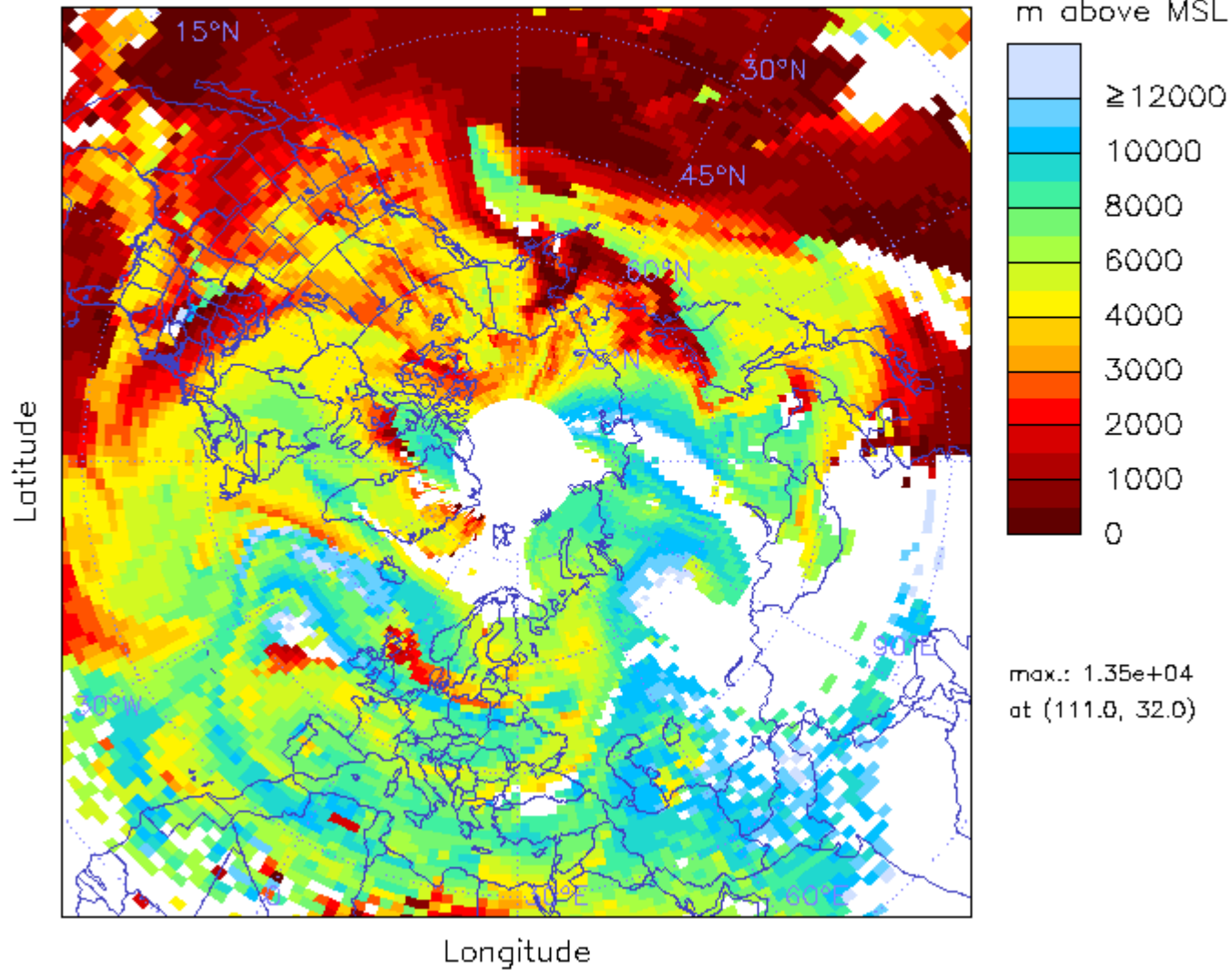


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 23 Mar

median height of distribution, 00:00 UTC 23 Mar

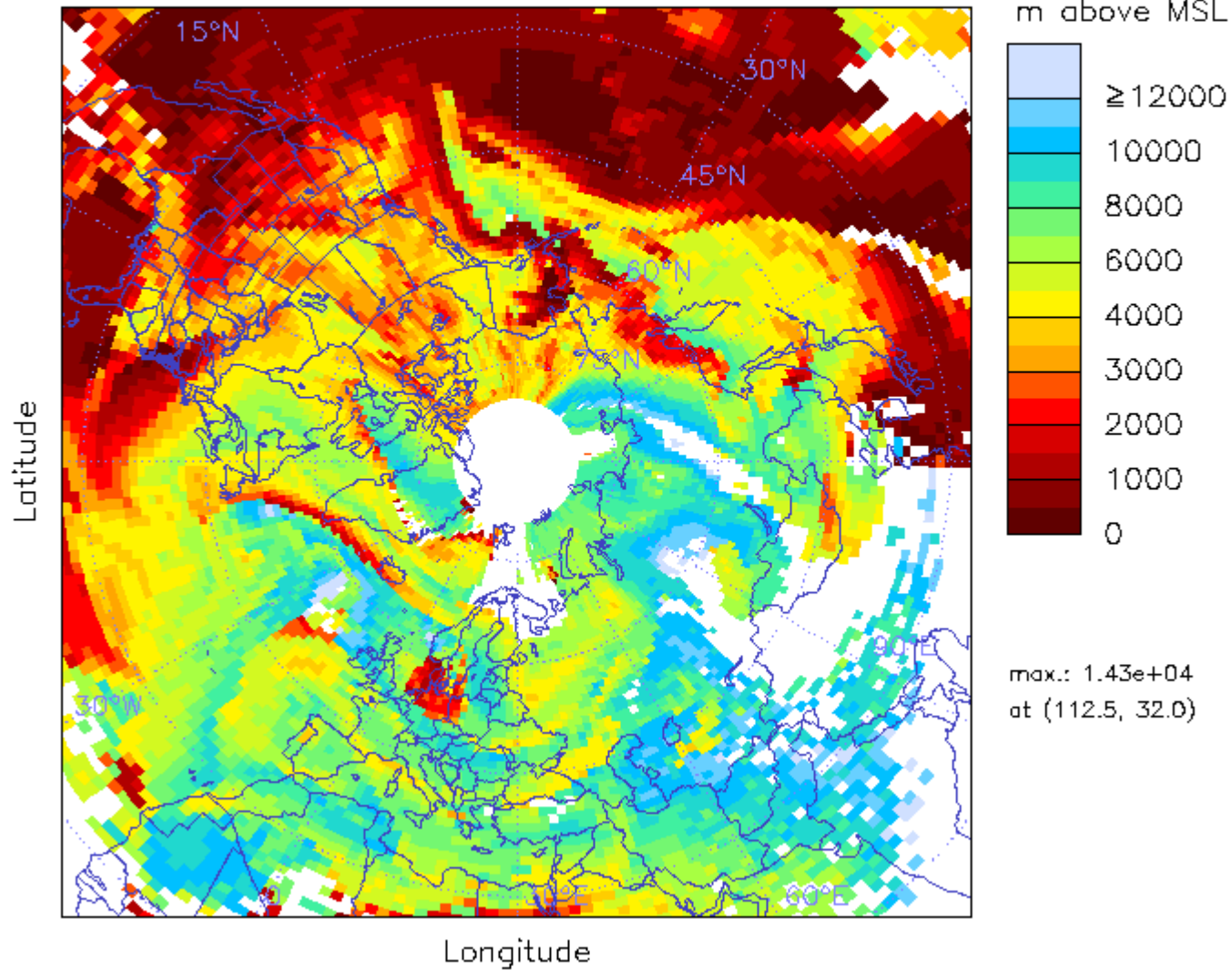


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 12 UTC 23 Mar

median height of distribution, 12:00 UTC 23 Mar

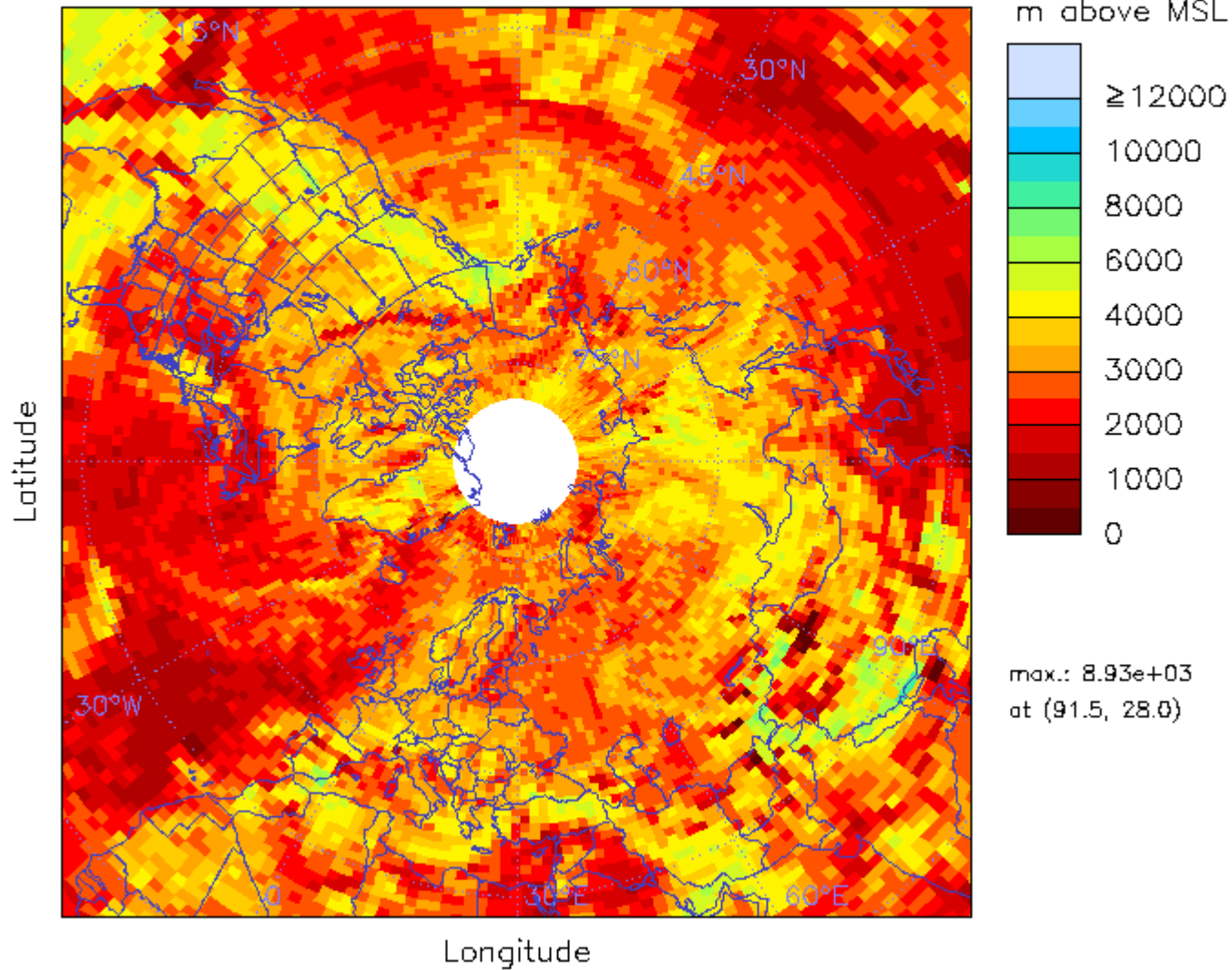


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 14 Apr

median height of distribution, 00:00 UTC 14 Apr



# Figures 4a,b

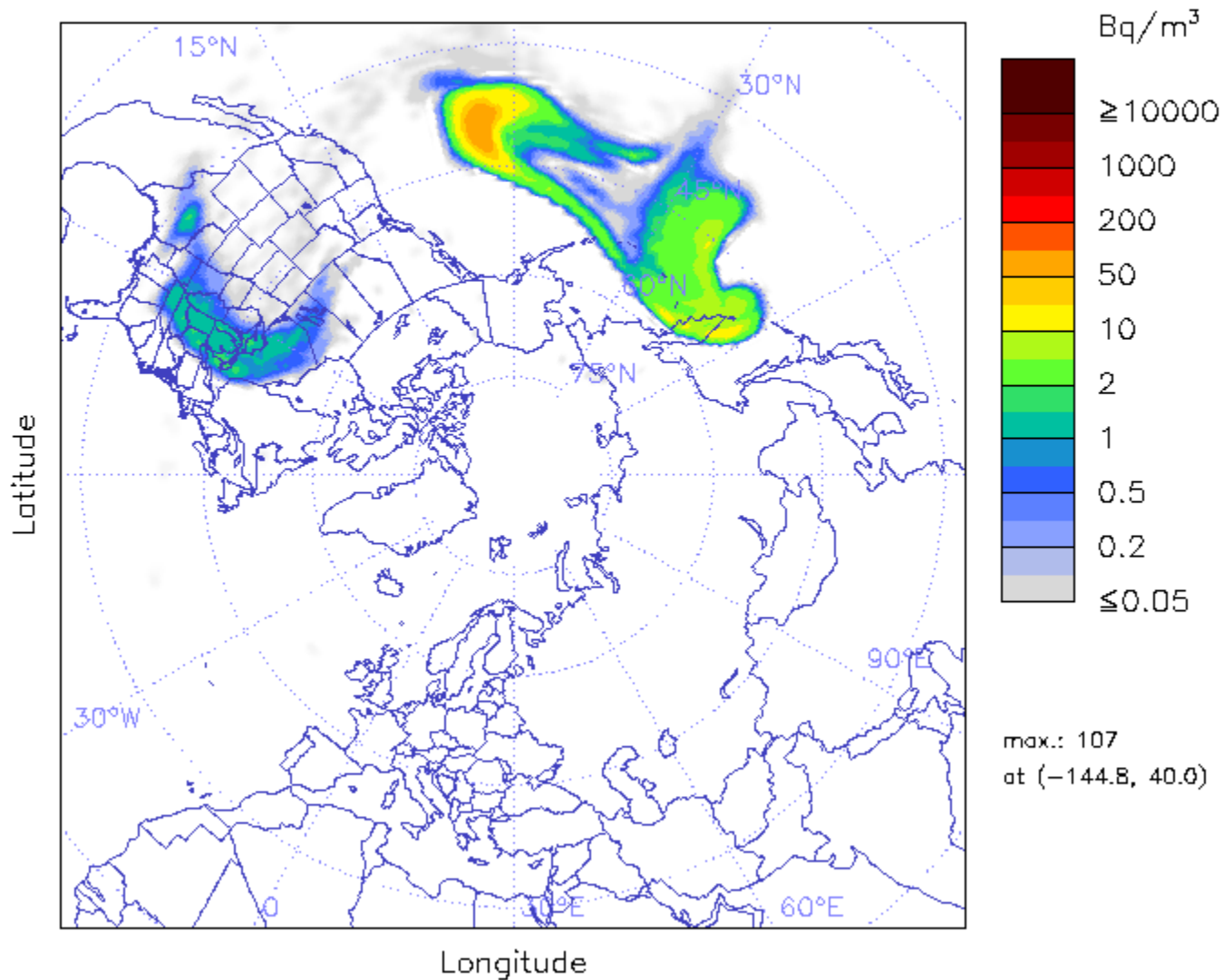
- Model-1 simulation of the evolution of the Fukushima Xe-133 plume in the UTLS, above 9 km. The color code indicates the maximum Xe-133 activity concentrations in the same vertical column segments as in Figures 2. Snapshots are shown only for: 17 March (a); 23 March; day of our first aircraft flight (b) .

NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 00 UTC 17 Mar

max. conc. 9000 to 15000 m, 00:00 UTC 17 Mar

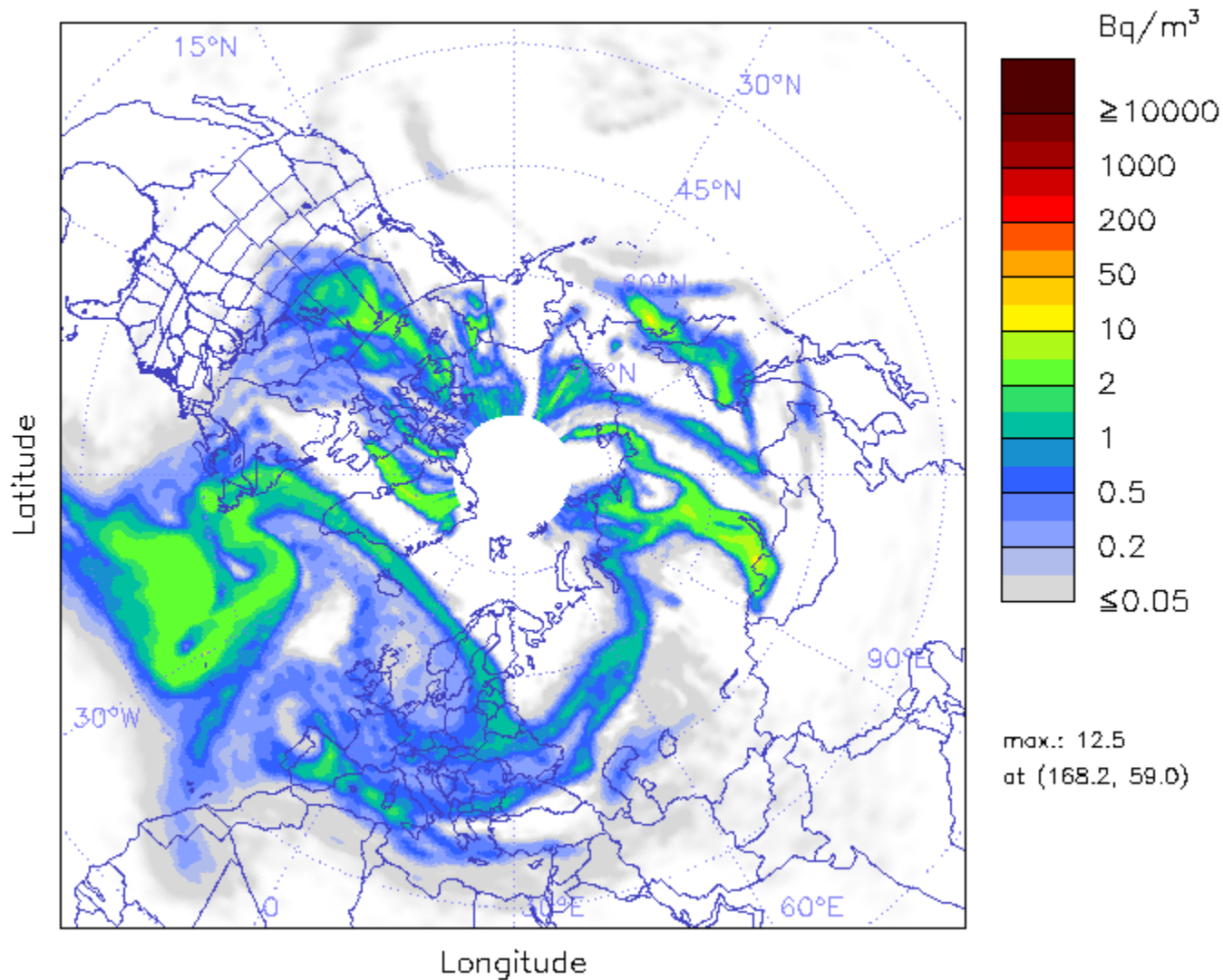


NOAA HYSPLIT model

X133 release starting 12:00 UTC 11.03.11

GDAS forecast init: 12 UTC 23 Mar

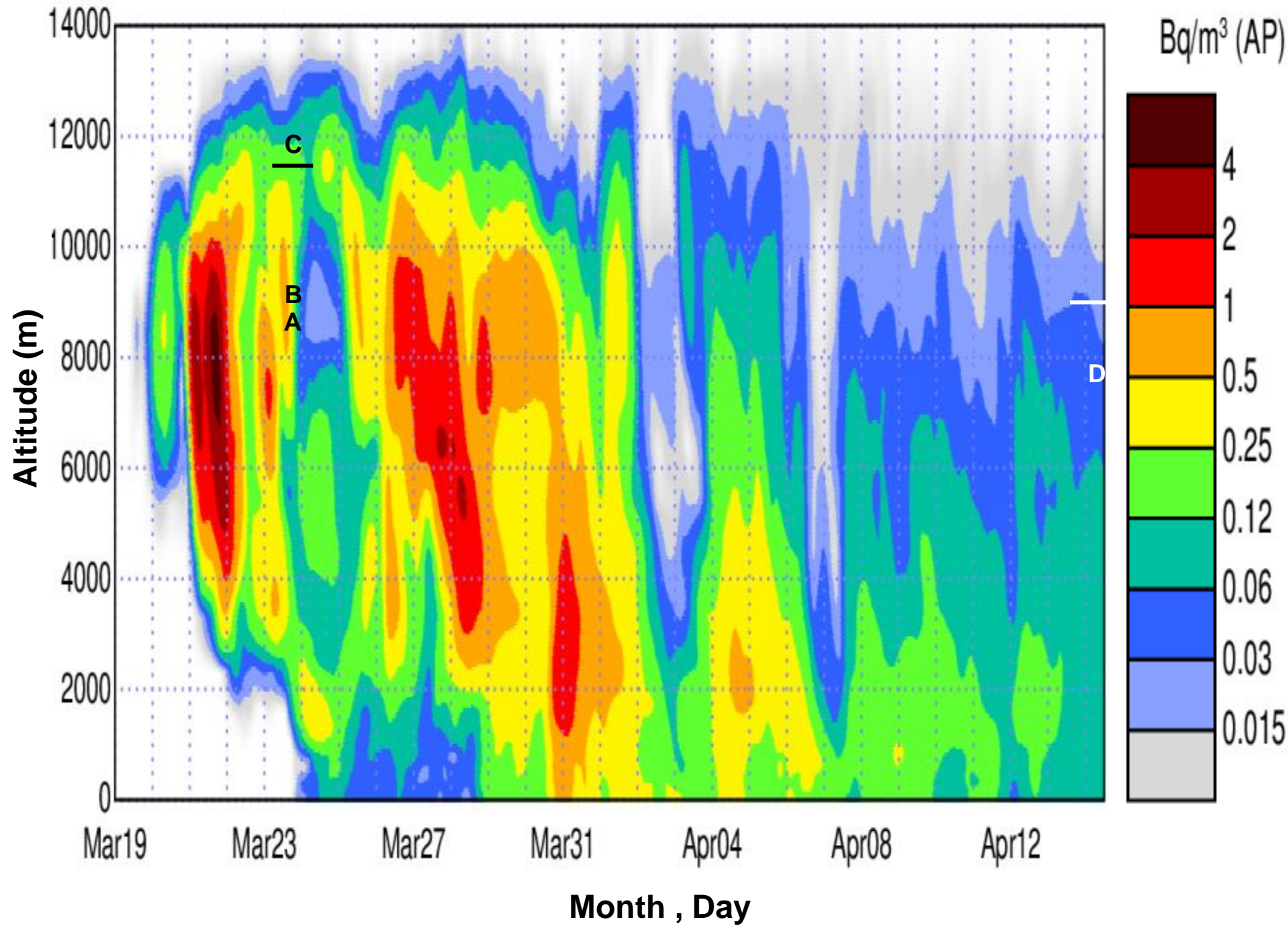
max. conc. 9000 to 15000 m, 12:00 UTC 23 Mar



# Figure 5

Altitude-time cross section of model run-1 Fukushima Xe-133 activity concentrations (unit: Bq/m<sup>3</sup> at ambient atmospheric pressure).





# Figure 6

Model-1 simulation of FD Xe-133 activity concentrations (24 hour averages) over South-Germany (48 N), for 3 selected height ranges (11.83-12.3 km, 8-9 km, 0-2 km above sea level). The two dashed curves have a decreasing trend resulting only from radioactive decay of Xe-133. The dashed curves are normalized respectively to the red and blue model-1 curves, after 31 March (when the FD plume was already spatially relatively homogeneous). Also shown are our airborne Xe-133 activity concentrations (green boxes), measured over South-Germany (around 48N).

