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Fukushima Xe-133 detection at the tropopause: Implications for very efficient na no-particle formation at the tropopause from lifted SO2 of East-Asian origin.

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Abstract

We report determined airborne measurements, at the tropopause, of the radioactive noble gas Xe-133, accidentally released by the mayor nuclear power plant disaster at Fukushima (mostl y in the period 11-15 March 2011). Our unique measurements, representing the first and so fa r 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 measurementsallowed us to use Xe-133 as an ideal tracer for WCB induced transport of grou nd-level air to the tropopause, followed by intercontinental high-altitude transport. Our measu rement and our accompanying air transport model simulations reveal that parts of the Fukushi ma 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 lif t (within about 12 hours) to about 10 km altitude. Hereafter, the plume was carried by high-al titude winds across the North-Pacific, North-America, and the North-Atlantic. During this int ercontinental transport, the plume experienced additional moderate ascent. Already on 20 Ma rch the plume reached the Upper troposphere and lowest stratosphere over Central-Europe, w here our first research aircraft flight took place on 23 March 2011. Our second flight took pla ce on 14 April when the, meanwhile considerably diluted, plume had already spread over larg

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

Scientific background

The tropopause (TP) region, and particularly the TIL (Tropopause Inversion Layer), locate d just above the the TP) represents an interesting atmospheric region, offering ideal condition s for SO2 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 S O2 input into the TIL is not well known. This is particularly true for SO2 input via anthropog enic SO2 lift from ground-level SO2 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 thicknes varies with season and latitude. I mportant details of TIL formation are as yet not fully understood.

Potential sources of SO2 present in the TIL are: airplanes cruising in or in the vicinity of the TIL, whose engines release SO2 as a combustion product of fuel sulfur; volcanic SO2 input; lift of SO2 from ground-level SO2 sources.

Lift of SO2 from ground-level SO2 sources to the TIL is not well known. The efficiency of vertical air transport from the planetary boundary layer of mayor anthropogenic SO2 emission regions, which are located mostly in middle latitudes of the northern hemisphere, is only poorly known. Also poorly known is heterogeneous SO2 loss by cloud processes, occurring during lift. Relevant heterogeneous SO2 loss processes include washout, rainout, and liquid-phase conversion to sulfate.

The strongest ground-level fossil fuel combustion related SO2 releases to the atmosphere are presently occurring in middle-latitude East-Asia (East-China/ Korea/ Japan; hereafter: CK J region), followed by Europe and eastern North-America (c.f. EDGAR archive; new Paper). Efficient transport from the JKC region ground level SO2-sources to the tropopause and the T IL 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 n orthern 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 sim ulations 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 SO2-rich pollution plumes in the middle and upper troposphere, which according to our previous model simulations, had been lifted by East-Asia n WCBs. However, so far no such measurements have been reported for the tropopause regio n and the TIL. The first airborne mass spectrometric SO2 measurements in the entire TIl-altit ude 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-1 33, 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 rad io-nuclides. Among these, was also the radioactive noble gas Xe-133, which was released pre ferably 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 tr acer since it is chemically inert and therefore represents a tracer, which is not affected by che mical 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 require d for WCB mediated transport in the UTLS (Upper Troposphere/Lower Stratosphere) from F ukushima to Europe. Importantly, the Xe-133 lifetime, at and above the tropopause, is also ap roximately comparable to the time scale for OH-induced SO2 conversion in the (e-folding lif etime of SO2: about 5-10 days). Hence, the concentration ratio lifted SO2/Xe-133 should re main 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 f uel 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 fue l rods, the fuel rod temperature rapidly increases and as a consequence Xe-133 is readily relea sed from the fuel rods. Interaction of water with the hot fuel rods may lead to catalytic H2 ge neration, which may lead to an H2/O2 explosion. Such an explosion may damage the pressur e vessel, resulting in uncontrolled Xe-133 release to the atmosphere. In order to avoid an H2/ O2 explosion, the pressure vessel may occasionally be vented in a controlled way, leading to rapid release of H2 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 H2/O2 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 Mar ch 2011) lifted to the tropopause. Hereafter, the lifted plume experienced intercontine ntal transport to Europe, where it arrived on 20 March 2011.

b) During intercontinental transport, some additional ascent took place, even to the TIL.

c) The initial large-scale lift was, without doubt, induced by a West-Pacific WCB (War m conveyor Belt) associated with a pronounced cyclone.

d) Radioactive radiation doses received by passengers and crewmembers of airliners, pe netrating the Fukushima plume at typical cruise altitudes, are small compared to the ra diation doses aviation passengers receive from galactic cosmic radiation.

e) The most important conclusion from our investigation is: We have found an efficient t ransport path connecting the region with the strongest WCBs with the region of the lar gest fossil fuel combustion related pollution sources. This is particularly true for the f ossil fuel combustion generated gas SO2, 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:

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Figure 1

Title

Authors

Institutions

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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

D-ADLR
HALO

Figures 3a-h

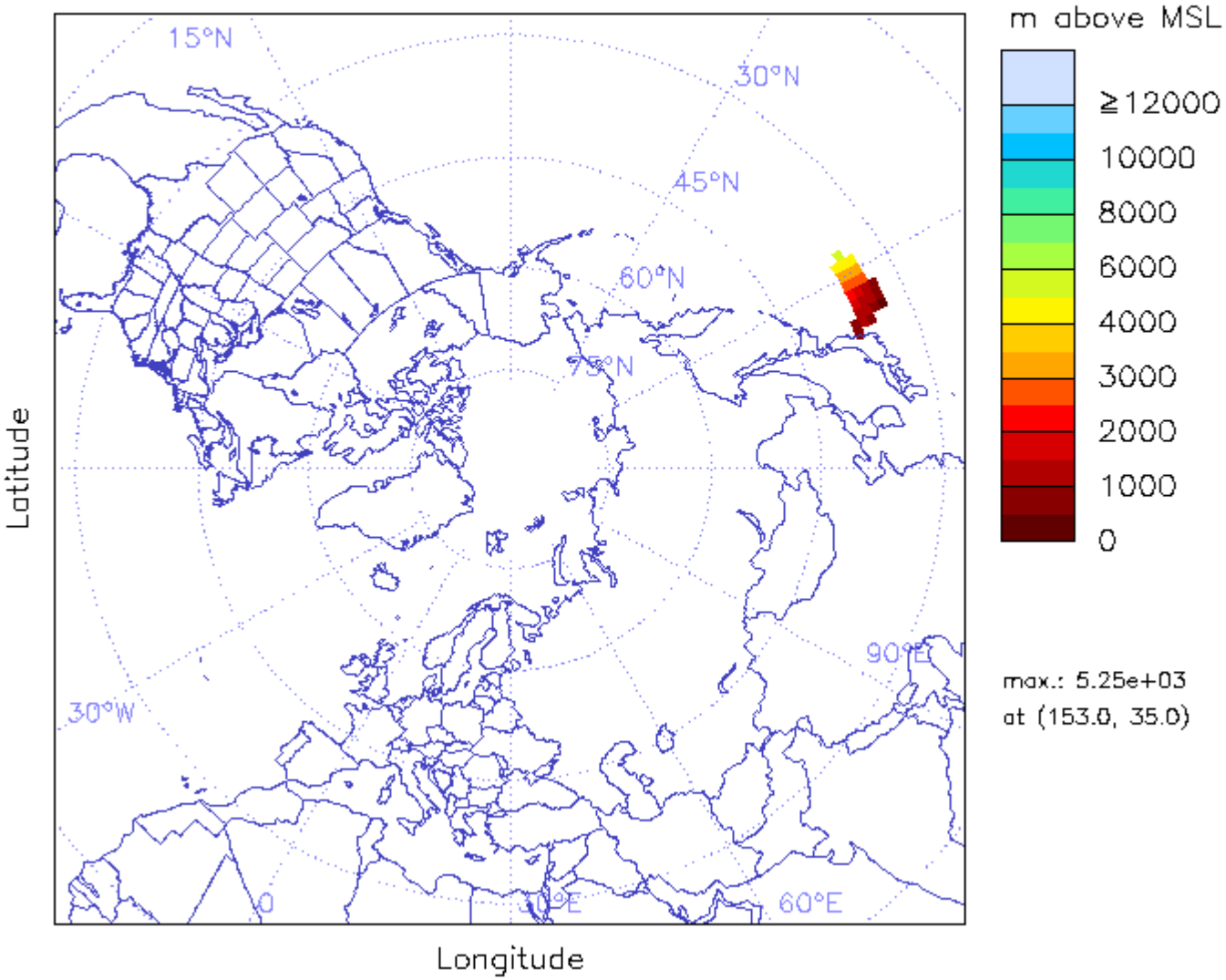
- Model-1 simulation of the evolution of the Fukushima Xe-133 plume. Plotted is a time-series of snapshots of the median-height of the plume for the period 12 March 2011 – 14 April 2011

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

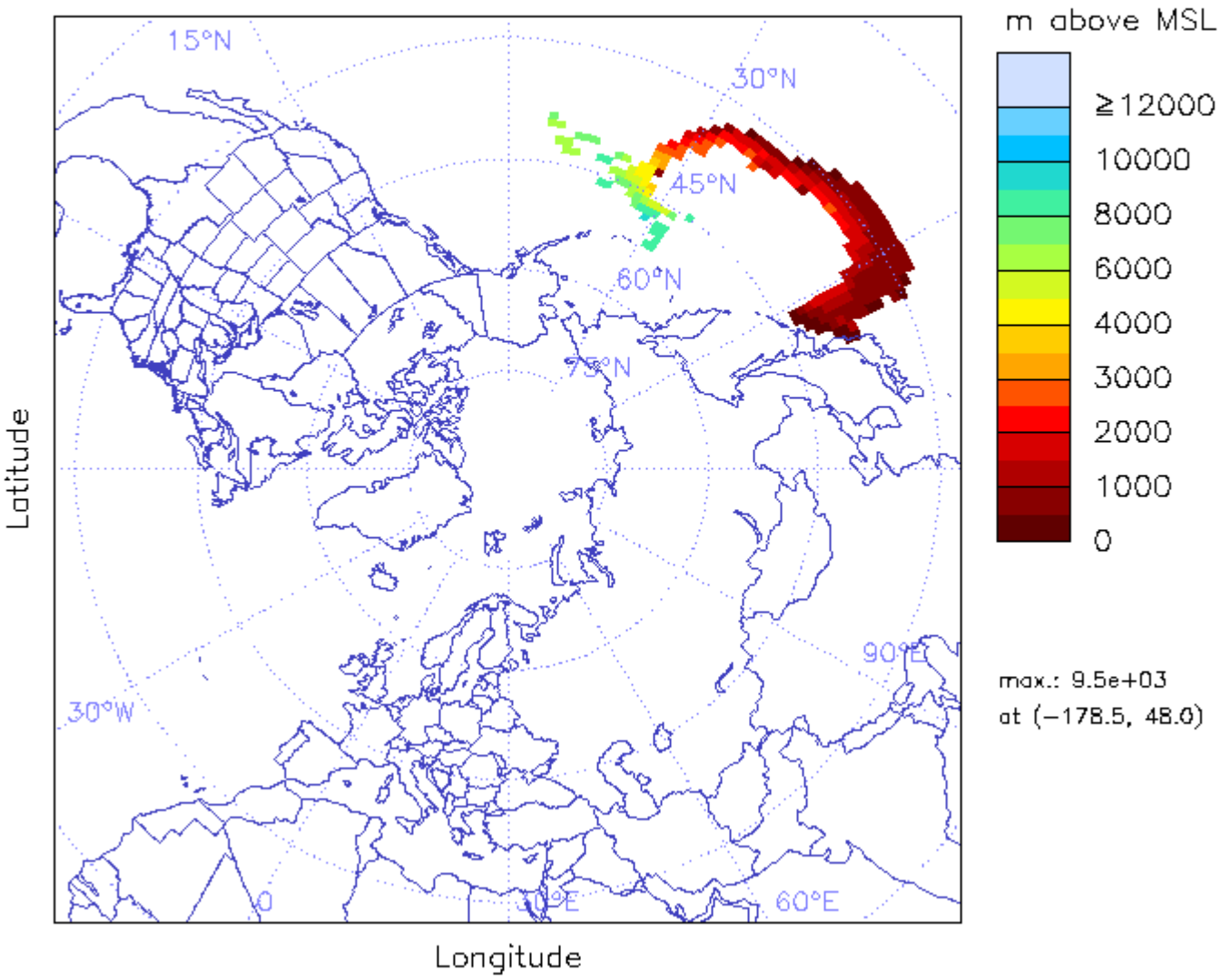


NOAA HYSPLIT model

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

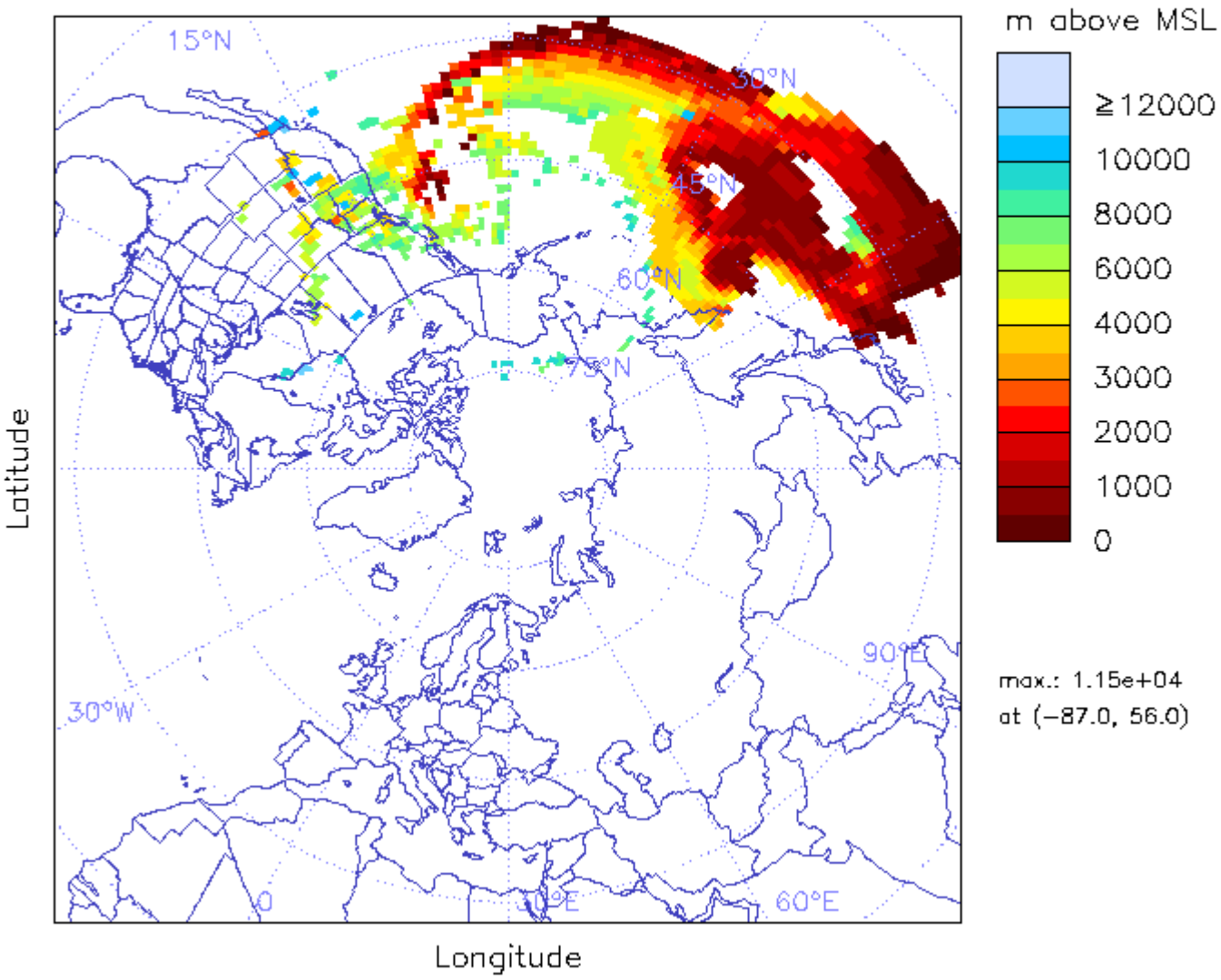


NOAA HYSPLIT model

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

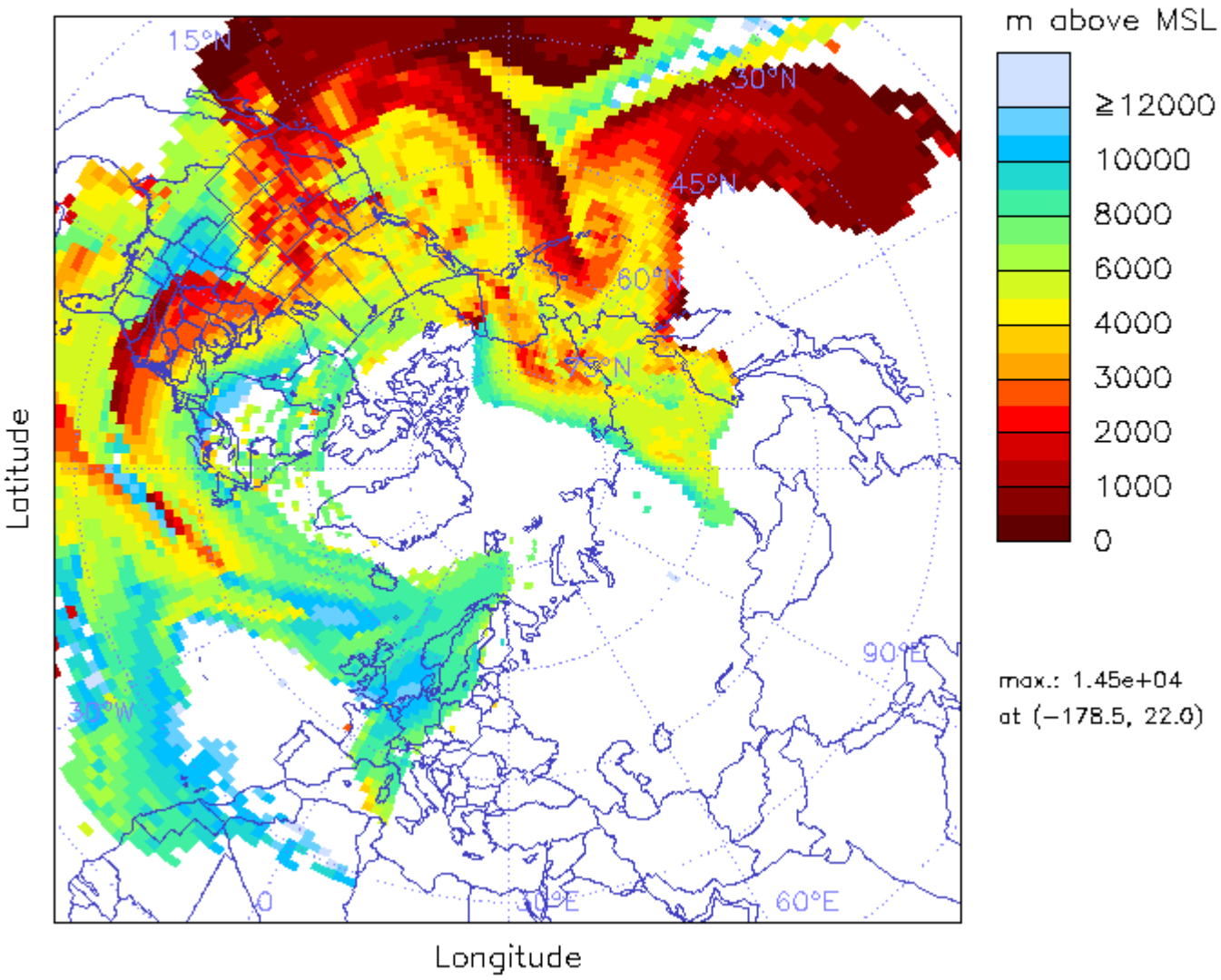


NOAA HYSPLIT model

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

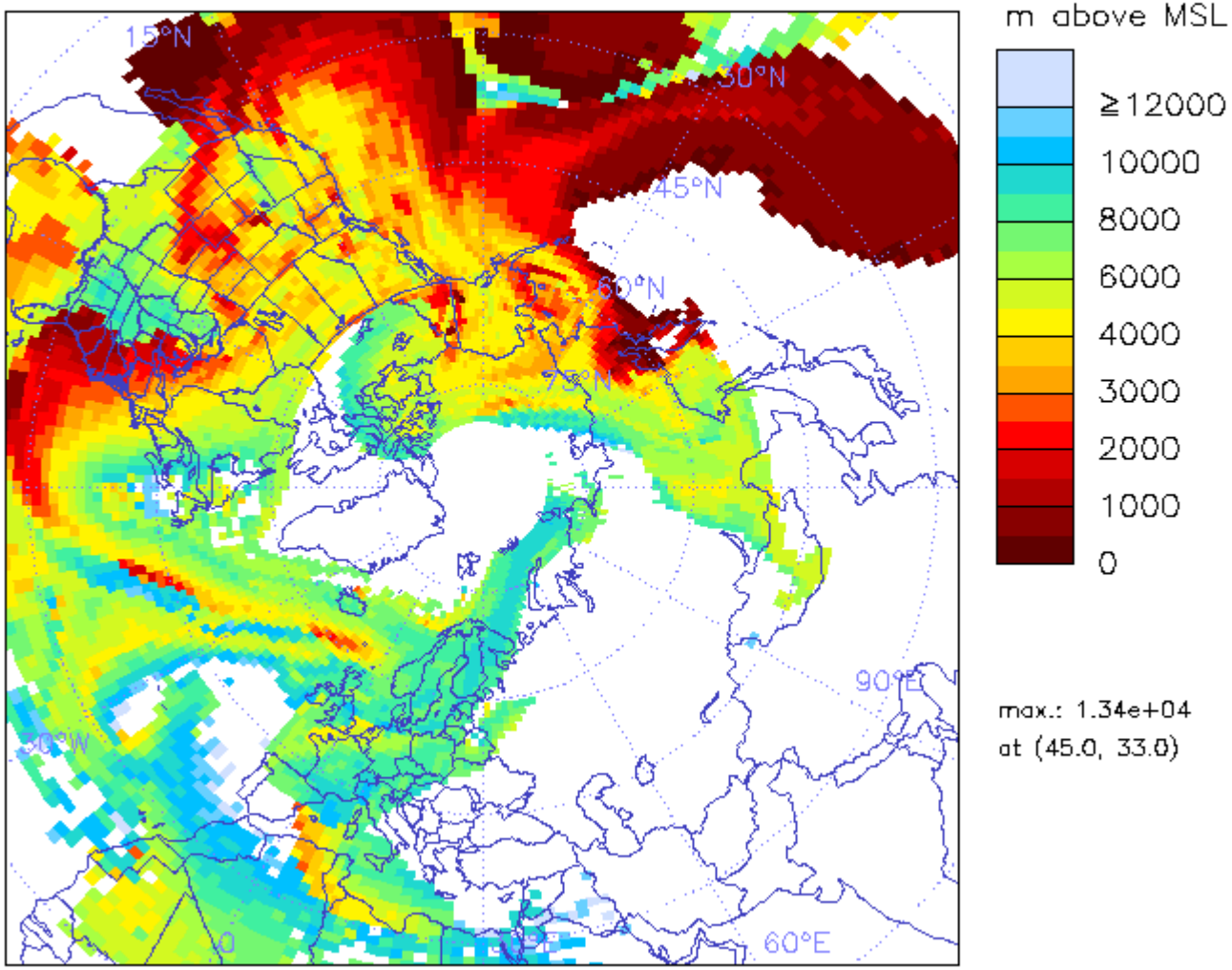


NOAA HYSPLIT model

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



m above MSL

≧12000
10000
8000
6000
4000
3000
2000
1000
0

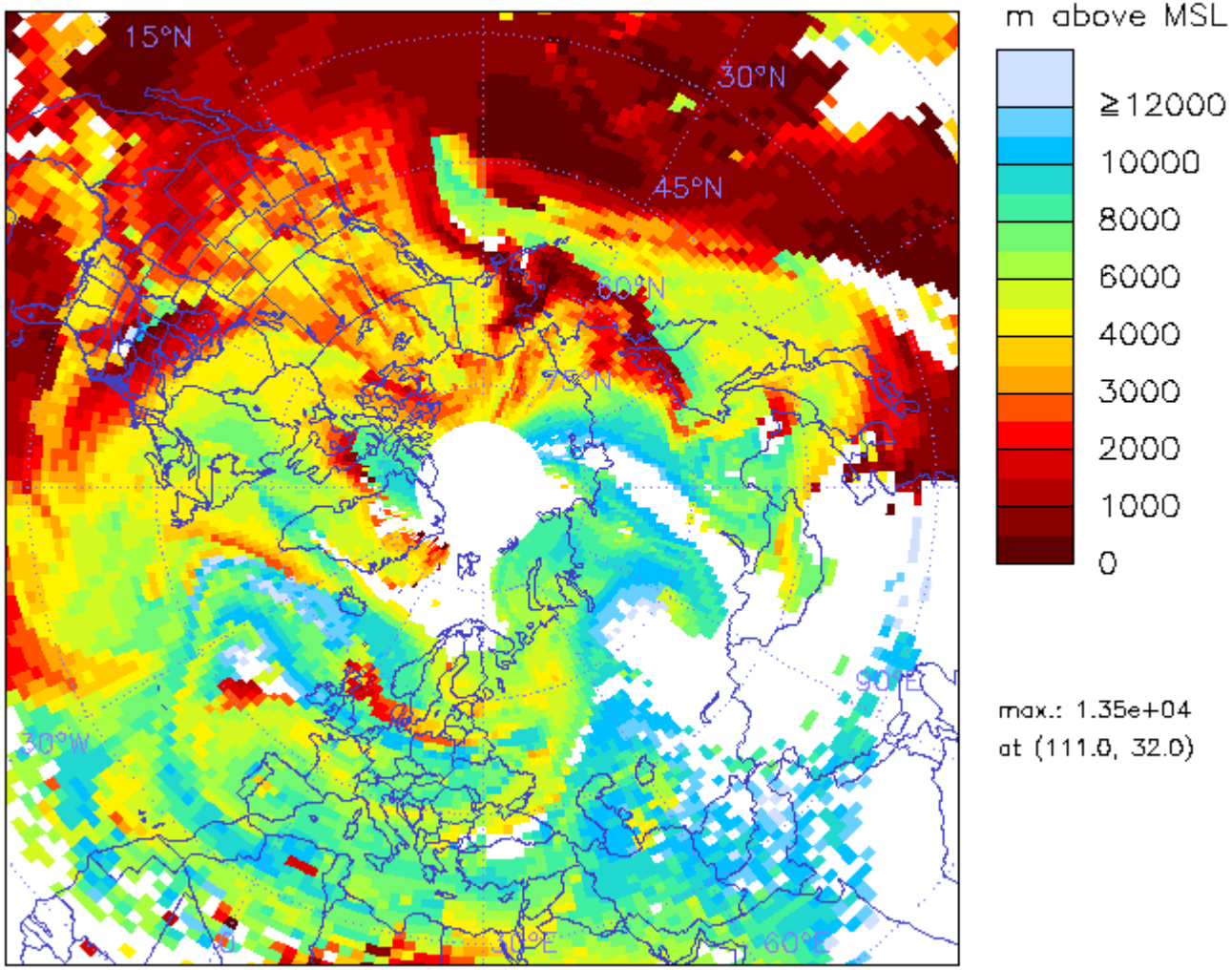
max.: 1.34e+04
at (45.0, 33.0)

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



m above MSL

≧12000
10000
8000
6000
4000
3000
2000
1000
0

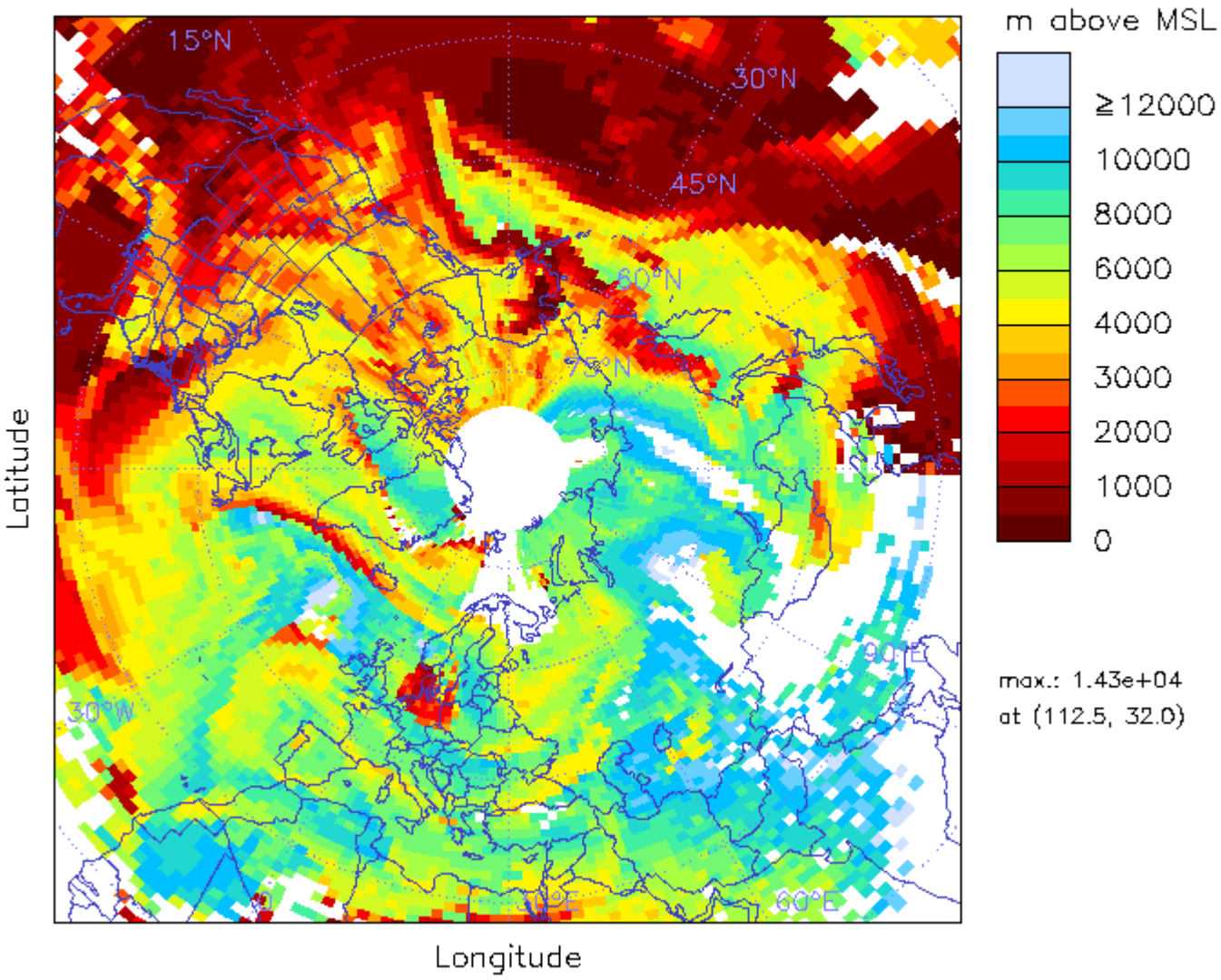
max.: 1.35e+04
at (111.0, 32.0)

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

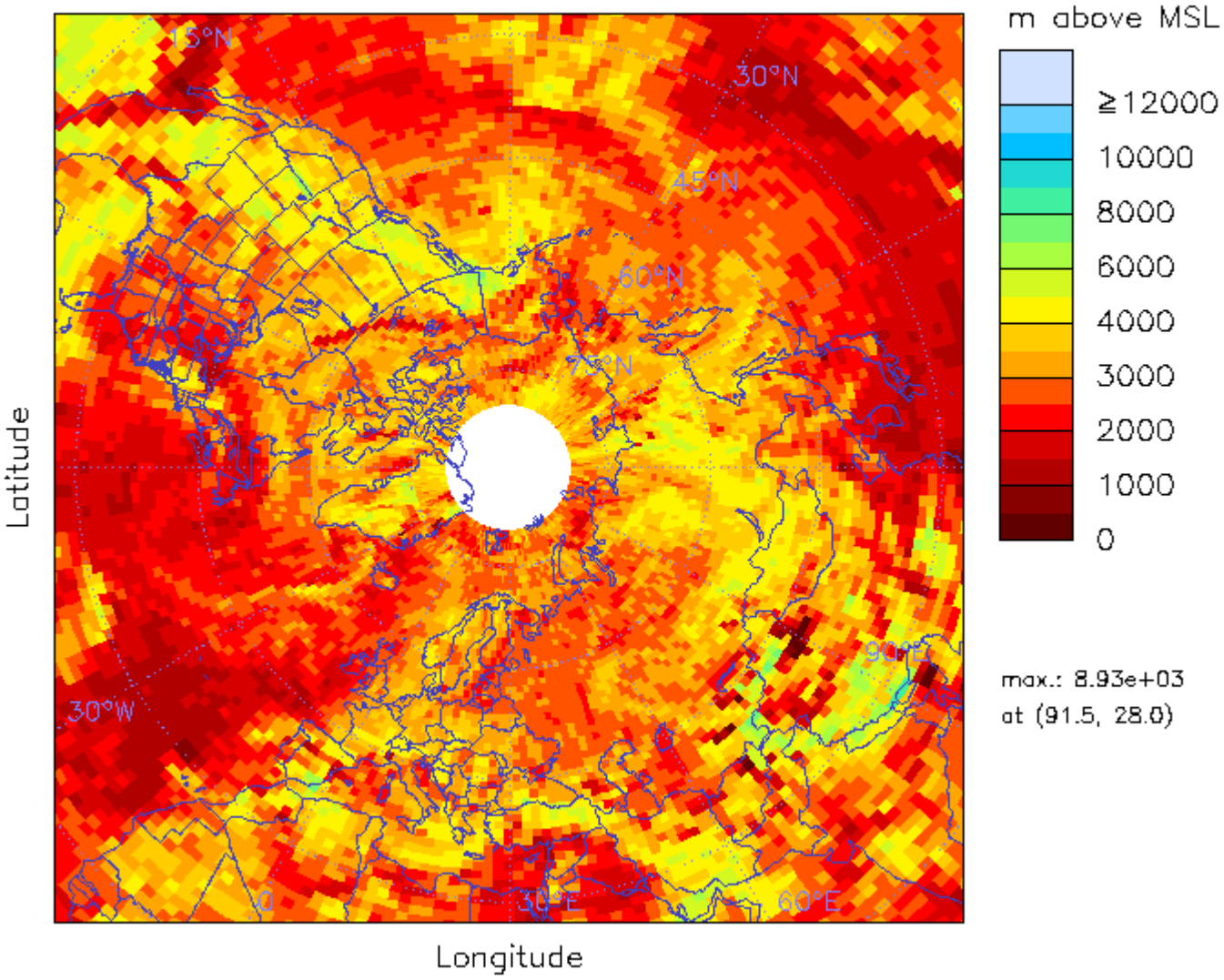


NOAA HYSPLIT model

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



m above MSL

≧12000
10000
8000
6000
4000
3000
2000
1000
0

max.: 8.93e+03
at (91.5, 28.0)

Figures 4a,b

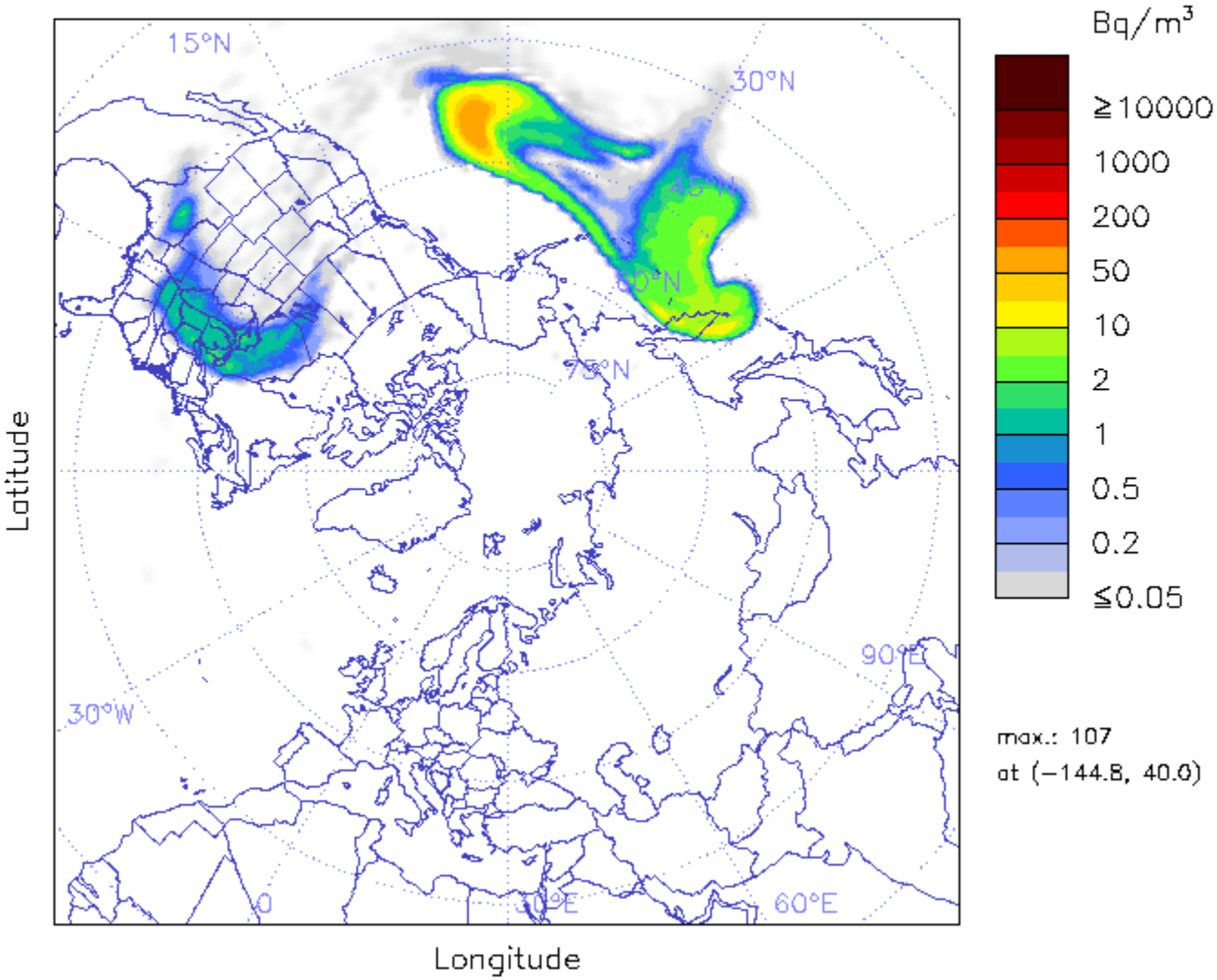
- Model-1 simulation of the evolution of the Fukushima Xe-133 plume in the UTLS, above 9 km. The color code indicates the maximum Xe-133 activitiy concentrations in the same vertical column segments as in Figures 2. Snapshots are shown only for: 17March (a); 23 March; day of our first aicraft flight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

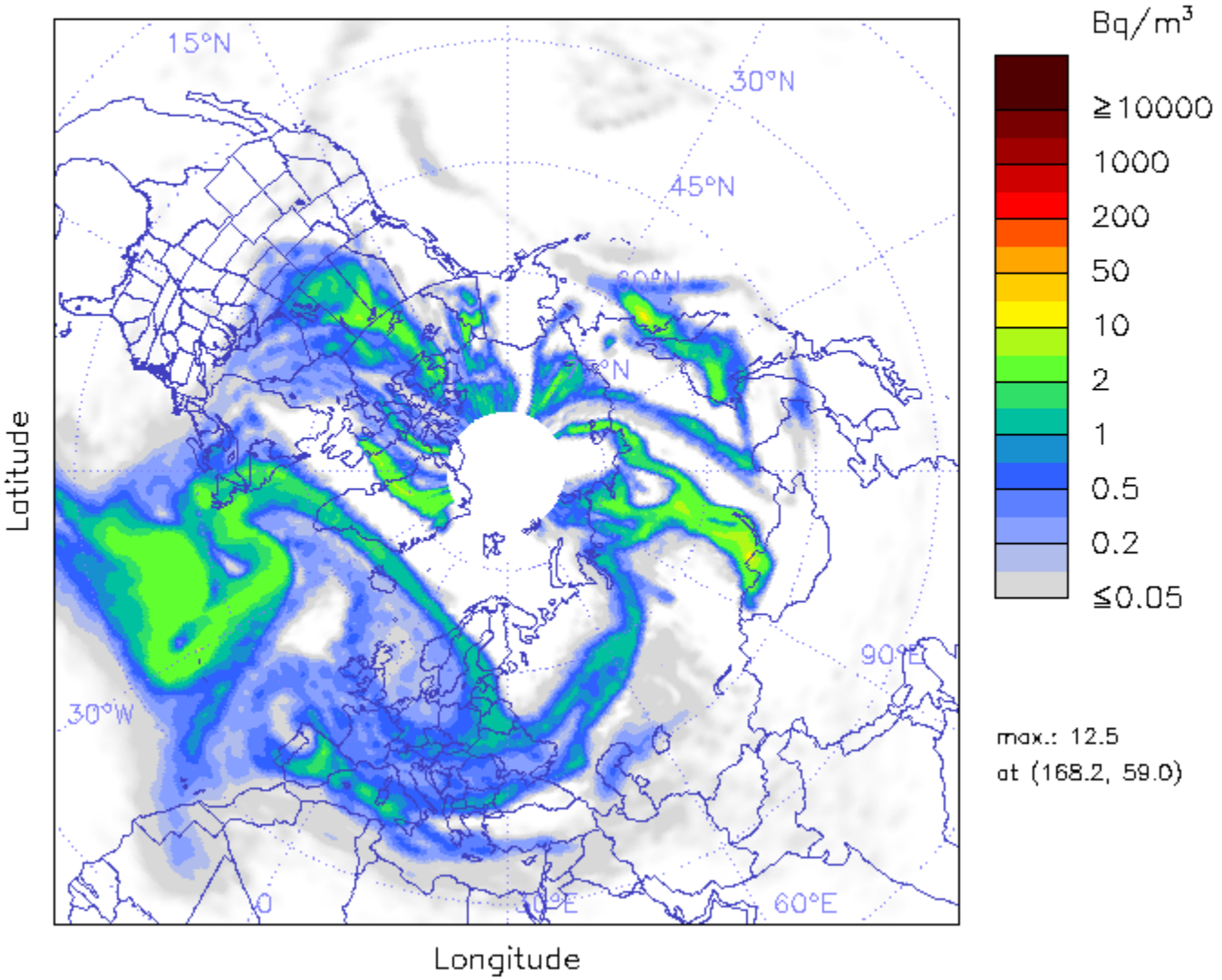


Figure 5

Altitude-time cross section of model run-1 Fukushima Xe-133 activity concentrations (unit: Bq/m3 at ambient atmospheric pressure.

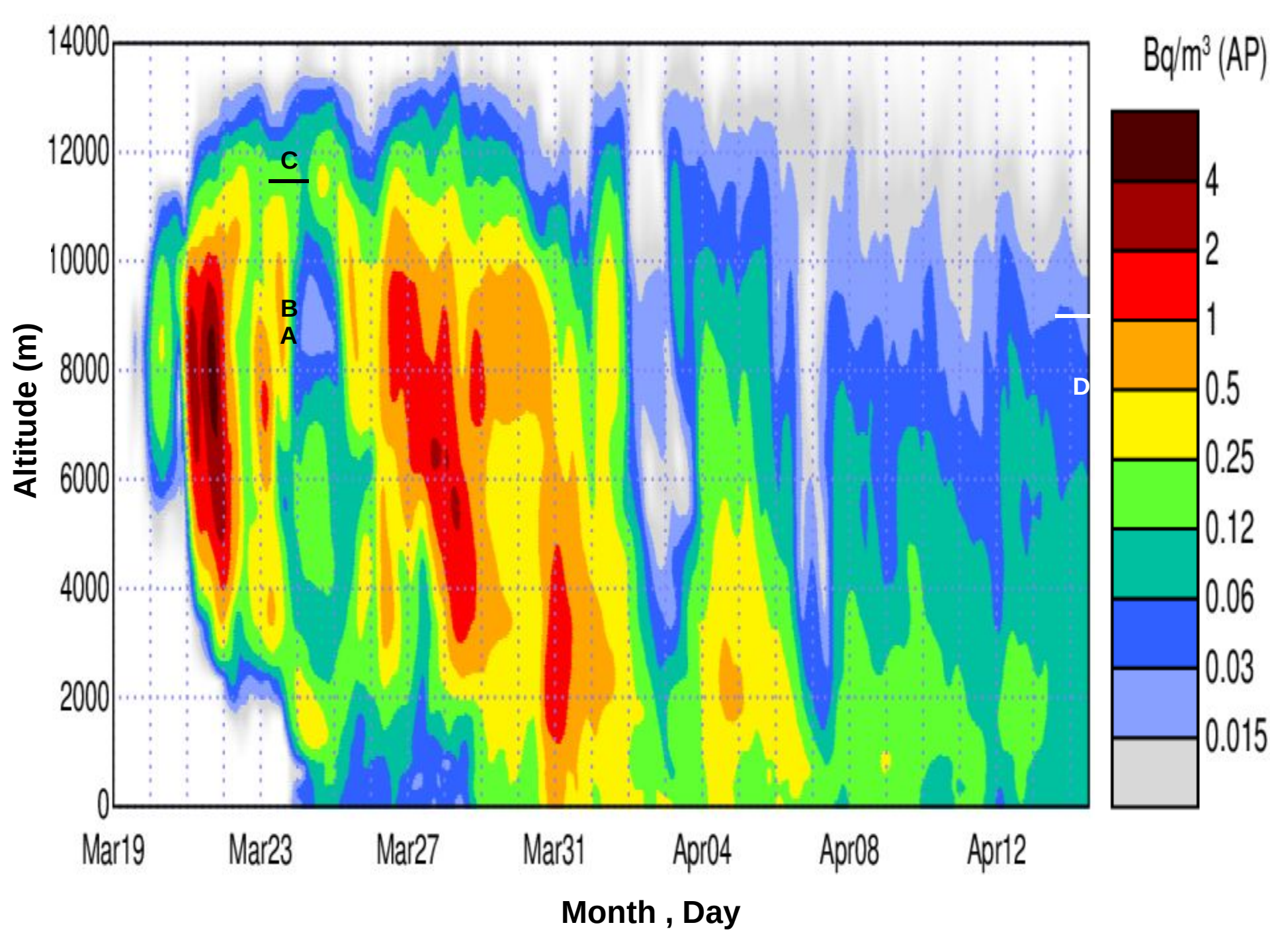

Bq/m3 (AP)
4
2
1
0.5
0.25
0.12
0.06
0.03
0.015
Altitude (m)
14000
12000
10000
8000
6000
4000
2000
0
C
B
A
D
Mar19
Mar23
Mar27
Mar31
Apr04
Apr08
Apr12
Month , Day

Figure 6

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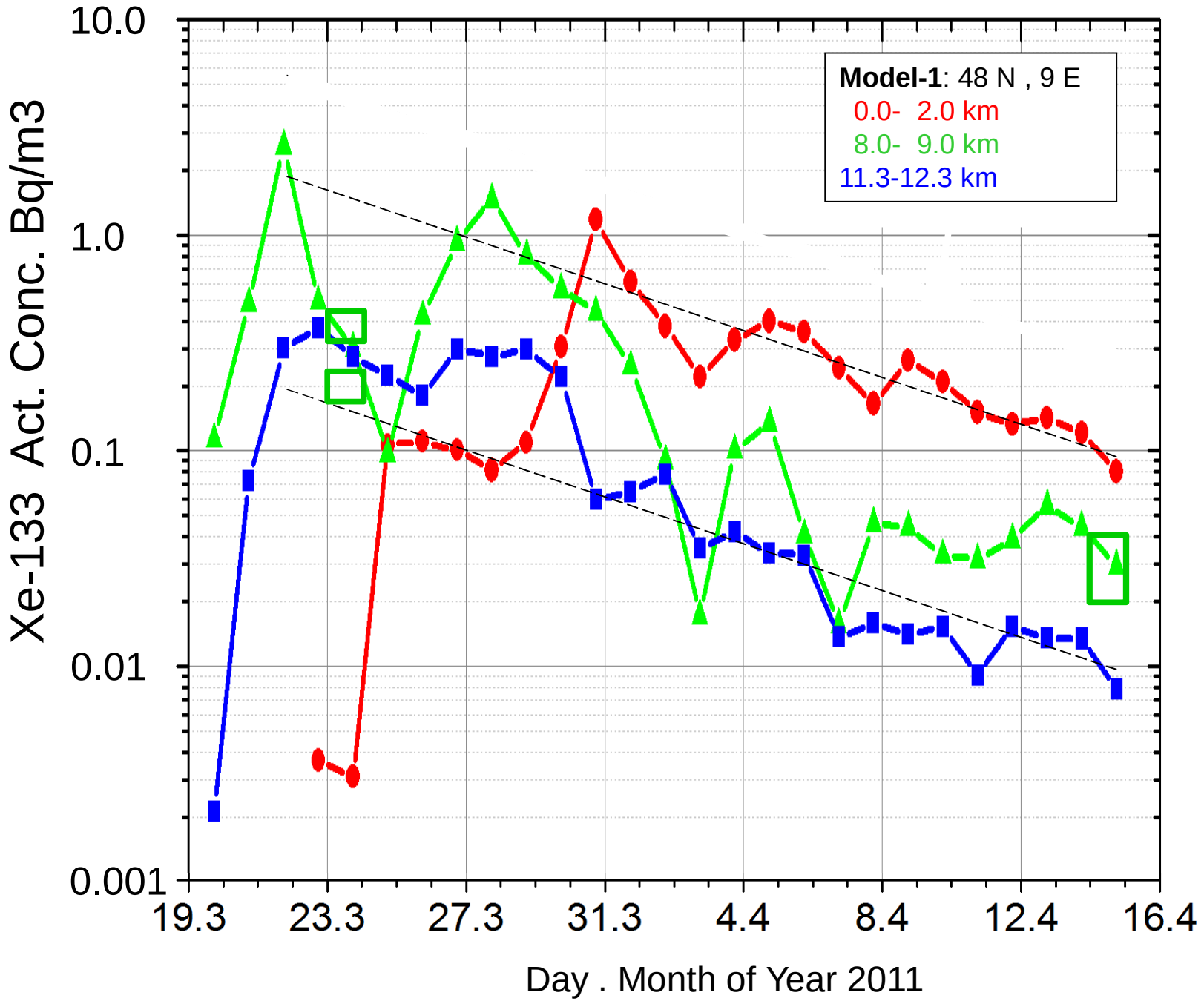
Model-1 simulation of FD Xe-133 activity concentrations (24 hour averages) over South-Germany (48 N), for 3 selected height ranges (11.83-12.3 km, 8-9 km, 0-2 km aove sea level). The two dashed curves have a decreasing trend resulting only from readioactive decay of Xe-133. The dashed curves are normalized 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).


Model-1: 48 N , 9 E
0.0- 2.0 km
8.0- 9.0 km
11.3-12.3 km
Xe-133 Act. Conc. Bq/m3
10.0
1.0
0.1
0.01
0.001
19.3
23.3
27.3
31.3
4.4
8.4
12.4
16.4
Day . Month of Year 2011