Detection of ^{133}Xe from the Fukushima nuclear power plant in the upper troposphere above Germany

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Abstract

After the accident in the Japanese Fukushima Dai-ichi nuclear power plant in March 2011 large amounts of radioactivity were released and distributed in the atmosphere. Among them were also radioactive noble gas isotopes which can be used as tracers to probe global atmospheric circulation models. This work presents unique measurements of the radionuclide ^{133}Xe from Fukushima in the upper troposphere above Germany. The measurements involve air sampling in a research jet aircraft followed by chromatographic xenon extraction and ultra-low background gas counting with miniaturized proportional counters. With this technique a detection limit of the order of 100 ^{133}Xe atoms in liter-scale air samples (corresponding to about 100 mBq/m^3) is achievable. Our results proof that the ^{133}Xe-rich ground level air layer from Fukushima was lifted up to the tropopause and distributed hemispherically. Moreover, comparisons with ground level air measurements indicate that the arrival of the radioactive plume in Germany in high altitude is several days earlier than on ground.

Keywords: Fukushima, Reactor accident, Low-level gas counting, Ultra-low background, Radioxenon, Xenon-133.

1 Introduction

One of the strongest earthquakes in Japanese history happened at the east coast of Honshu Island on March 11, 2011 and was followed by a destructive tsunami. This triggered a series of accidents in the nearby Fukushima Dai-ichi nuclear power plant [1] which caused a release of large amounts of radionuclides [2, 3, 4, 5] among them also the radioactive xenon nuclide ^{133}Xe. The total ^{133}Xe source term was estimated to be between 12 EBq and 19 EBq [5, 6, 7]. Such a strong release opens up the possibility to probe global atmospheric circulation models using ^{133}Xe as a tracer. Since xenon is a noble gas it is inert and rarely reacts with other elements. Thus, it is superior to other commonly used tracers which require profound understanding of atmospheric chemistry, washout processes and rain. Moreover, its lifetime of 7.57 days

[8] is comparable to the time scale for intercontinental transport in the upper troposphere. In this work we present the detection of airborne ^{133}Xe in the upper troposphere above Germany after the Fukushima event. A second paper [9] deals with the atmospheric aspects and implications of the ^{133}Xe measurements as well as with the interpretation of simultaneously measured trace gases and aerosols. For comparison with data of ground level measurements in Europe obtained with iodine and cesium radionuclides we refer to [10].

After first information about the nuclear accident became public we prepared an aircraft campaign on short notice. The rationale of our aircraft measurements was to investigate pollution of the upper troposphere, particularly the tropopause region, by pollutants released from fossil fuel combustion in the region of North-East China, Korea and Japan. In fact, this region represents the strongest SO_2-source region
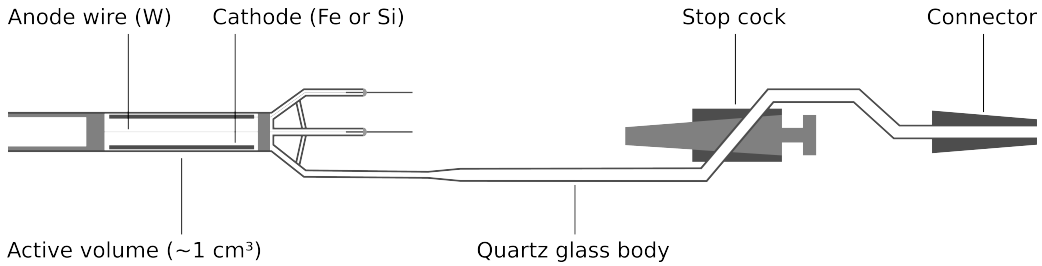
Figure 1: Sketch of the miniaturized ultra-low background proportional counter used for the ^{133}Xe measurements.

worldwide. Since SO_2 is an important precursor of potentially climate active atmospheric aerosol particles, above mentioned region became a matter of particular interest at the Deutsche Luft- und Raumfahrtzentrum (DLR) [9, 11, 12, 13].

Samples were taken during two flights with the Falcon research jet aircraft [14] of DLR. The first flight on March 23, 2011 was aiming for the interception of the Fukushima ^{133}Xe plume in the upper troposphere above Germany, shortly after its expected arrival on March 21/22, 2011. Due to strict air safety regulations only rather small (1 liter) and passive air samplers were approved in the short time before the first flight. The second flight on April 14, 2011 aimed to probe the highly diluted Fukushima plume in the upper troposphere. Here larger samples of about 10 liters could be taken. Model simulations conducted by the Institut für Physik der Atmosphäre of DLR prior to the flights, predicted that upper tropospheric ^{133}Xe activity concentrations above Germany would be around 1 Bq/m^3 during the first flight and about 10 times less during the second flight. Thus, extremely sensitive ^{133}Xe detection techniques in the laboratory are required to observe the Fukushima signal in liter-scale samples.

2 ^{133}Xe measurements with miniaturized proportional counters

Miniaturized ultra-low background proportional counters (see Figure 1) were developed at the Max-Planck-Institut für Kernphysik to detect few atoms of ^{71}Ge in the Gallex solar neutrino experiment [15]. They were also used for ^{133}Xe studies in Gallex [16] and later to detect low levels of various radioactive noble gas isotopes in the astroparticle physics experiments Borexino [17], Gerda [18] and Xenon [19]. Due to their low background and low energy threshold they are ideally suited to detect gaseous radioactive isotopes with good sensitivity. The key technology is a dedicated processing of gas samples before loading them into the counter. It includes removal of trace impurities which disturb the performance of the proportional counter (e.g. oxygen, humidity, ...) as well

as separation of unwanted radioactive impurities. Finally, the size of any gas sample has to be reduced without losing target isotope and without introducing contaminations to fit into the active volume of the miniaturized counter (~ 1 cm^3). The sample preparation is done by means of a dedicated gas handling and counter filling line made from glas. Such a system was developed in the framework of astroparticle physics experiments and as indicated in [20] it may be used for xenon processing.

The main challenge is the xenon separation at high acceptance from the air samples. This is done by pumping the sample over a 0.6 gram activated carbon column held at -186 °C by immersing it in liquid argon. While most of the oxygen and nitrogen is pumped away xenon is efficiently stopped in the column. Subsequently, the sample is transferred to the top of a gas chromatography column by heating the activated carbon column and cooling the gas chromatography column at liquid nitrogen temperature. Helium is used as a carrier gas to run the sample through the gas chromatography column which is filled with Chromosorb 102 [21]. Nitrogen, oxygen, carbon dioxide, xenon and radon elute from such a column in the listed order. The temperature is varied step-wise from liquid nitrogen temperature to -110 °C and finally to -30 °C to achive a good separation of all components. A big challenge is ^{222}Rn in the sample eluting shortly after xenon. Radon is not visible in the chromatogram which is recorded by a thermal conductivity detector with limited sensitivity. Thus, it is necessary to reject the late fraction of the xenon which might not be fully separated from ^{222}Rn. This causes losses between five and ten percent for a single run. They are quantified via a natural xenon carrier which is added to the sample beforehand. Xenon has no long-lived radioactive isotopes, therefore the carrier which is several years old does not introduce new background. Together with 10 % of the quenching gas methane the xenon is used as a counting gas for the proportional counters which are operated at atmospheric pressure. The radon issue will be discussed
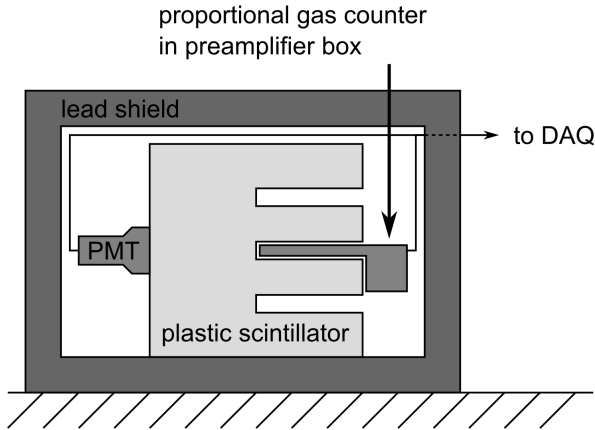
Figure 2: Sketch of the gas counting setup with passive shield and active plastic scintillator veto system.

Figure 3: Spectrum of a ^{133}Xe standard for efficiency calibration. The bump around 5 keV is due to admixed ^{131m}Xe, which is present at the few percent level in the ^{133}Xe standard.
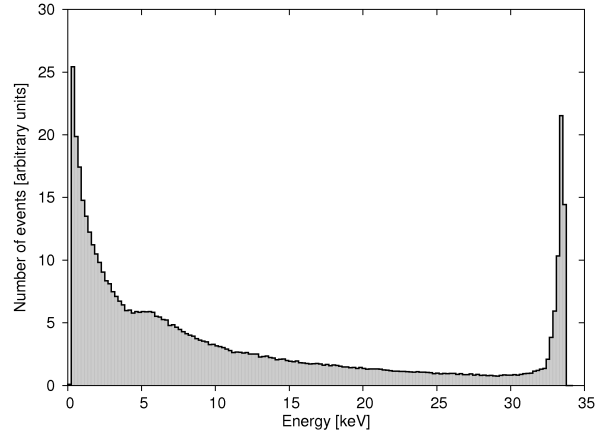
in more detail in section 6.

The energy calibration is done by illuminating the proportional counters with cerium X-rays according a technique which was developed for the GALLEX experiment [22]. The cerium X-rays excite xenon atoms and generate a discrete low-energy line spectrum in the proportional counter. The energy to channel allocation is done by fitting a second order polynomial to the positions of the three peaks that appear at 1.1 keV, 5.0 keV and 9.7 keV. For some runs the full absorption peak centered at 34.6 keV was included in the fit and it was confirmed that a second order polynomial provides a good description up to that energy. Another peak at 0.3 keV was not considered, because it was not always clearly resolved due to its vicinity to the threshold.

3 Counting system

The counting system (see Figure 2) to read out the proportional counters is located at the subterranean Low-Level-Laboratory of the Max-Planck-Institut für Kernphysik in Heidelberg, which is at a depth corresponding to about 15 m of water equivalent. The counters are placed in a plastic scintillator veto system which provides almost 4-π coverage to reject cosmic muons. The plastic scintillator itself is embedded in a 15 cm lead shield.

^{133}Xe decays with a lifetime of 7.57 days by beta disintegration with a Q-value of 427.4 keV (see Table 1). 99.12 % of all decays populate a 81 keV isomeric state (lifetime 9.1 ns) which de-excites with 37 % probability by emission of a gamma quant and in the remaining cases by internal conversion [8]. In the proportional counters we measure the energy deposition of emitted electrons and of X-rays from simultaneous de-excitation of the atomic shell. Typically, only a small fraction of the released energy is deposited in the miniaturized counters, thus a low energy threshold is required. At around 150 eV the efficiency of our data aquisition system drops down significantly. To avoid threshold effects a conservative 500 eV cut was applied in the data analysis. The 81 keV gamma ray might trigger an unwanted veto signal in the plastic scintillator. Thus, the counters were directly surrounded by a $\sim$ 5 mm lead shield to which a box containing the frontend electronics is connected. Signals are read out by a Flash - Analog to Digital (FADC) converter board (Struck SIS3301) at 100 MHz sampling rate and 14 bit resolution. The risetime of a proportional counter pulse is defined by the time difference between 10 % and 90 % of its maximum amplitude. The plastic scintillator covers a surface of approximately 0.6 m^2 perpendicular to the muon flux resulting in a muon event rate of about 100 Hz. Each signal from the proportional counter that is not in coincidence with a plastic scintillator signal is recorded with the indication of timestamp, energy and risetime. ^{133}Xe-events have a risetime of $\sim$ 1 μs. A conservative risetime cut of 3 μs was applied to discriminate against a population of slow pulses which was present in some of the data.

4 Efficiency calibration

A commercial ^{133}Xe calibration standard was used to determine the detection efficiency of the proportional counters for ^{133}Xe. It was first measured with a high purity Germanium (HPGe) gamma spectrometer to identify unwanted radionuclides. Besides ^{133}Xe, the

Isotope	Lifetime τ	Q-value [keV]	Decay-mode
^{133}Xe	7.57 d	427.4	β-decay to excited levels
^{131m}Xe	17.21 d	163.9	Converted gamma transition
^{85}Kr	15.51 a	687.1	β-decay (0.43% to excited level)
^{222}Rn	5.52 d	5590.3	α-decay

Table 1: Selected decay data of relevant radionuclides taken from [8].

gamma rays from ^{131m}Xe and ^{85}Kr were visible in the spectrum indicating a contamination with these nuclides at the percent level. Figure 3 shows an energy spectrum of the calibration standard as recorded with one of the proportional counters.

Alpha-particles deposit a large amount of energy in the counter due to the high ionization density along their track. Thus, overflow channels are not considered in the analysis and an upper energy threshold of 27 keV is applied to remove these events.

Using also the selection criteria dicussed in section 3 good events are selected in a window between 0.5 keV and 27 keV and with a rise-time of less than 3 μs. To obtain the efficiency for ^{133}Xe the temporal decay curve of these events is fitted with two exponential decays for ^{133}Xe and ^{131m}Xe, respectively and a constant background component, which comprises mainly long-lived ^{85}Kr. In the fit the lifetimes of ^{133}Xe and ^{131m}Xe were fixed to the literature values given in Table 1. The obtained efficiencies for ^{133}Xe vary between 49 % and 56 % for the six counters in use. Such variation is expected due to geometrical differences of the counters resulting in different ratios of active to passive volumes.

5   Maximum likelihood analysis

The recorded events of all samples were analyzed with a maximum likelihood approach which was originally developed for data analysis in radiochemical solar neutrino experiments [23] . To estimate the initial number N of ^{133}Xe atoms in a given sample the probability density function of ^{133}Xe decays is used which is exponentially decreasing with the ^{133}Xe lifetime. In contrary, the background distribution is assumed to be constant in time with a rate b. The expected sources of background events are unvetoed muons and contaminations of the proportional counters with traces of radioanuclides (e.g. ^{210}Pb) with long lifetimes. Contributions from ^{131m}Xe, ^{133m}Xe and ^{135}Xe which were also released during the accident are negligible in the four samples of the first flight [7]. For the second flight data from the Bundesamt für Strahlenschutz (see section 7) suggest that the ^{131m}Xe fraction of the signal reaches 11 %. We checked that the inclusion of this component in the likelihood function does not

change the result for ^{133}Xe significantly. In particular, since the ^{131m}Xe also originates from Fukushima it slightly improves the significance for the detection of radioxenon from the accident. However, the efficiency for ^{131m}Xe was not calibrated, so we decided to ignore it in the analysis.

If t_i are the time stamps of the n recorded events and if T describes the time intervals of the total measurement, the resulting likelihood function $\mathcal{L}$ can be written as

$$\log \mathcal{L}(N,b) = -\int_T \left(\frac{N}{\tau} e^{-t/\tau} + b \right) dt +$$

$$+ \sum_{i=1}^n \log \left(\frac{N}{\tau} e^{-t_i/\tau} + b \right). \quad (1)$$

While the second part of this equation is derived by the probabilities of events occured at the time stamps t_i, the first part describes the likelihood of no observed events in between. Finally, $\log \mathcal{L}(N,b)$ is numerically maximised to get the best estimators $\hat{N}$ and $\hat{b}$ for the free parameters. In maximum likelihood theory, the statistical error of a fit-parameter a is estimated by varying the parameter around its best fit value $\hat{a}$ until

$$\log \mathcal{L}(\hat{a}) - \log \mathcal{L}(\hat{a} \pm \sigma_a) = \frac{1}{2} \quad (2)$$

while $\log \mathcal{L}(\hat{a} \pm \sigma_a)$ is maximised regarding the remaining free parameters. To consider a possible asymmetry of the error both sides of $\hat{a}$ are treated independently, however, the asymmetry turned out to be negligible in our case.

6   Radon correction

The ^{222}Rn concentration in ambient air fluctuates depending on local geological properties, on meteorological influences and on the altitude. Even in the upper troposphere it might not be negligbile with respect to the expected ^{133}Xe signal from the Fukushima plume [24]. Thus, special care was taken to separate radon from xenon during the gas chromatography procedure (see section 2). However, since the separation is difficult it cannot be excluded that a few radon
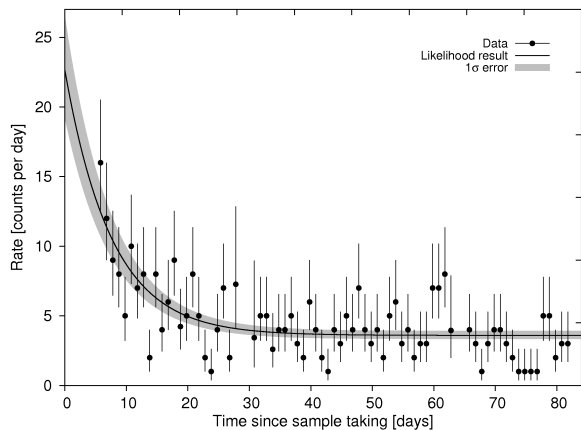
Figure 4: Temporal development of the measured decay rate per day for sample 'Flight A-1'. The excess due to presence of ^{133}Xe in the left part of the plot is clearly visible. The black curve shows the result and the 1 σ uncertainties (grey band) of the maximum likelihood analysis.

Sample	$\hat{N}$ [atoms]	$\hat{b}$ [cpd]
Flight A-1	72 ± 13	3.5 ± 0.3
Flight A-2	102 ± 18	8.8 ± 0.4
Flight A-3	< 37	5.6 ± 0.3
Flight A-4	27 ± 12	4.1 ± 0.4
Flight B-1	46 ± 16	5.1 ± 0.6
Flight B-2	61 ± 32	30.9 ± 1.4

Table 2: Results of the maximum likelihood analysis for the six samples. The errors are 1-σ statistical uncertainties and the upper limit is given at 90 % confidence level.

atoms entered the proportional counter. The lifetime $\tau = 5.52$ d [8] of ^{222}Rn is in the range of ^{133}Xe, therefore radon can not be treated as a part of the constant background b in the likelihood analysis.

To investigate a possible radon contamination of the samples a delayed coincidence analysis was performed: The ^{222}Rn decay chain includes ^{214}Bi with its short-lived progeny ^{214}Po (lifetime $237\,\mu$s [8]). Thus, a ^{214}Bi-^{214}Po (BiPo) coincidence event is characterized by a β-decay of ^{214}Bi followed by an α-decay of ^{214}Po on a short time scale. With the measurement of a ^{222}Rn standard sample the detection of one BiPo event was calibrated to correspond to an average of 10.6 ± 3.7 radon events in the energy region of interest between 0.5 keV and 27 keV.

From the six measured samples five showed no BiPo event, while there were two BiPo events in the sample 'Flight A-1' (see section 7). Since the gas handling procedure was similar for all six samples we assume the contamination risk to be the same for all samples. Thus, the number of radon atoms entering the counter is expected to be Poissonian distributed. Using the quoted numbers above, a likelihood analysis of this scenario leads to a correction of the number N of ^{133}Xe atoms in each sample by $3.5^{+3.3}_{-2.3}$.

7 Results

In total six samples were drawn during the two flights. The first flight took place on March 23, 2011 and four samples were filled to evacuated stainless steel containers of one liter volume. On the second flight on

April 14, 2011 we combined five samples from various places in Northern Germany and five samples from various places in Southern Germany to obtain two larger samples of about 10 liters air each. All samples were taken inside the aircraft's cabin where a constant pressure of 800 mbar is maintained. The cabin air is permanently exchanged with outside air and a complete exchange takes only a few minutes.

All samples were counted in our gas counting setup for several months such that the constant background rate could be precisely determined after the decay of ^{133}Xe. As an example, Figure 4 shows the data from sample 'Flight A-1'. The result of the maximum likelihood analysis is given in Table 2. For each sample the number of initial atoms at the start of the measurement $\hat{N}$ and the constant background rate $\hat{b}$ as described in section 5 are given. Sample 'Flight B-2' suffered from an unexpected high background which cannot be explained easily. Five of the six samples show a signal above the decision threshold. For sample 'Flight A-3' an upper limit at 90% confidence level is given.

The fit results are corrected for the ^{222}Rn contamination and then converted into a ^{133}Xe activity at the time of sampling by using the measured ^{133}Xe detection efficieny of the six proportional counters. Also the losses as determined with the xenon carrier (see section 2) are taken into account. Finally, the corresponding ^{133}Xe activity concentration is calculated normalized to air at standard temperature and pressure (STP: 273.15 K and 10^5 Pa). Again the asymmetry of the 1 σ uncertainties is ignored since it turned out to be negligible. The final results are shown in Table 3.

The ^{133}Xe concentration in ambient air is usually well below 50 mBq/m^3 with only a few timely restricted exceptions [25]. Consequently, we can conclude that in the first flight on March, 23 2011 a clear evidence of ^{133}Xe from the radioactive plume of Fukushima Dai-ichi in the upper troposphere above Germany was observed. The evidence for the second

Sample	Coordinates	Altitude [km]	^{133}Xe activity [mBq/m^3 (STP)]
Flight A-1	48°N, 11°E	8.1	540 ± 110
Flight A-2	48°N, 11°E	9.2	950 ± 180
Flight A-3	50°N, 11°E	9.2	< 310
Flight A-4	51°N, 11°E	11.8	210 ± 110
Flight B-1	Northern Germany	8.5	42 ± 16
Flight B-2	Southern Germany	8.5	63 ± 36

Table 3: Measured ^{133}Xe activity concentrations and coordinates / altitude of the sampling position.
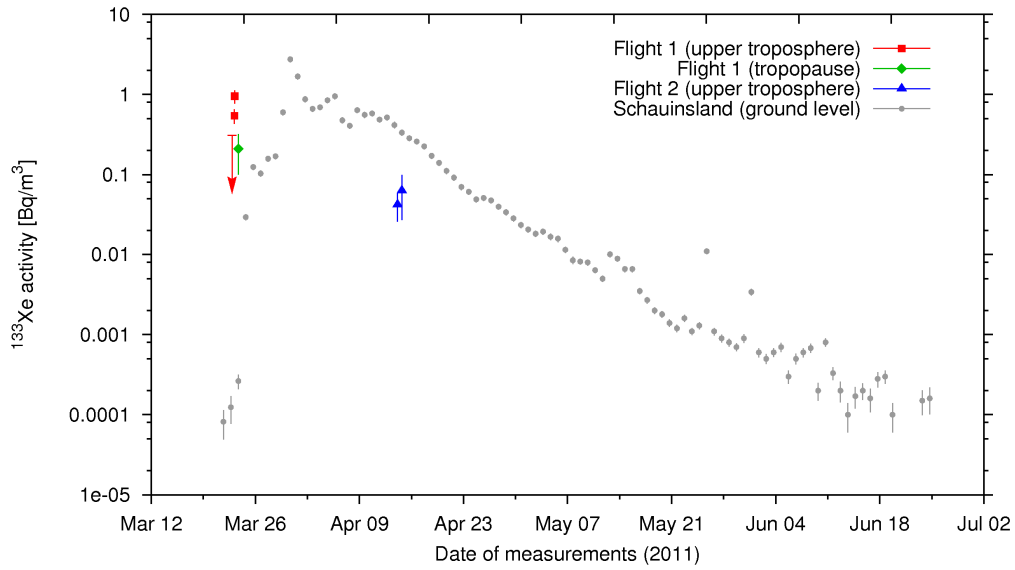
Figure 5: Comparison of the measured ^{133}Xe concentration during the flights and in ground level air recorded by the Bundesamt für Strahlenschutz on the Schauinsland mountain in Southern Germany.

flight about three weeks later is weaker, since ^{133}Xe has decayed and is further diluted. Nonetheless, our results are still above the usual ambient concentration indicating that we were still detecting the emission from Fukushima.

The Bundesamt für Strahlenschutz (BfS) operates a station for monitoring the environmental radioactivity at the Schauinsland mountain in Southern Germany. As Radionuclide Station 33 it is part of the International Monitoring Network (IMS) of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) [26, 27] and a SPALAX noble gas system [28] is installed to monitor continuously the activity concentration of radioactive xenon in ground level air. It is based on a fully automated sampling of large air samples, purification, xenon concentration and detection by gamma ray spectroscopy with a high purity Germanium detector. With the system measurement on 24 hours samples of four xenon radioiso-

topes (^{133}Xe, ^{133m}Xe, ^{131m}Xe and ^{135}Xe) in ambient air are performed. For ^{133}Xe the minimum detectable activity concentration lies around 450 μBq/m^3 (STP). Note the air sample size of typically 60 m^3 (STP) which is about 10^5 times larger than the air samples taken during the flights. In Figure 5 the results from the Schauinsland station in the time after the Fukushima accident are shown together with the results from the two flights. On ground level the first indication of the arrival of ^{133}Xe from Fukushima is visible on March 24, 2011. However, ^{133}Xe activity concentrations at the 1 Bq/m^3 level are detected for the first time on March 29, 2011.

The relatively high ^{133}Xe activity concentrations measured during the first flight on March 23, 2011 in the upper troposhpere show that the arrival of the Fukushima plume in high altitudes was earlier than on ground level. Also remarkable is the evidence for ^{133}Xe traces in the tropopause (sample Flight A-

4, green diamond in Figure 5). Both observations indicate that air masses from the Japanese regions were quickly lifted to high altitudes. For more information of atmospheric implications and comparisons with ground-level measurements the reader is referred to [9]. The ^{133}Xe activity concentrations in Flight 2 were significantly lower than simultaneous ground level air measurements at Schauinsland. This may be explained by settling of the heavy xenon in the atmosphere and is also confirmed by model calculations presented in [9].

8 Conclusions

We reported on a new technique successfully combining ^{133}Xe detection using miniaturized proportional counters with air plane sampling campaigns. In particular, we showed that small air samples of liter scale (STP) are sufficient to achieve a competitive detection limit in the range of 100 mBq/m^3 (STP). This is due to both, the ultra-low background rate of the applied proportional counters which is in the range of few events per day at 0.5 keV threshold and the highly efficient and contamination-free gas sample preparation procedure.

Our measurements reveal that a part of the Fukushima ^{133}Xe plume was lifted to the upper troposphere and to the tropopause. There it was carried by fast high-altitude winds to Europe. The earlier arrival time in Germany at high altitude compared to ground level was unambiguously demonstrated by a comparison with data from the CTBT Radionuclide Station 33 of the Bundesamt für Strahlenschutz on the Schauinsland mountain. We also got evidence for ^{133}Xe traces from the Fukushima accident in the upper troposphere during our second flight on April 14, 2011, although radioactive decay, dilution and settling of xenon reduced the ^{133}Xe activity concentration significantly.

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