An aerosol particle containing enriched uranium encountered in the remote upper troposphere

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A B S T R A C T

We describe a submicron aerosol particle sampled at an altitude of 7 km near the Aleutian Islands that contained a small percentage of enriched uranium oxide. ^{235}U was 3.1 ± 0.5% of ^{238}U. During twenty years of aircraft sampling of millions of particles in the global atmosphere, we have rarely encountered a particle with a similarly high content of ^{238}U and never a particle with enriched ^{235}U. The bulk of the particle consisted of material consistent with combustion of heavy fuel oil. Analysis of wind trajectories and particle dispersion model results show that the particle could have originated from a variety of areas across Asia. The source of such a particle is unclear, and the particle is described here in case it indicates a novel source where enriched uranium was dispersed.

1. Introduction

Aerosol particles can travel great distances in the atmosphere, so any sampling of such a particle represents the possibility of dispersal of uranium into the environment. Particulate matter containing uranium can originate from sources such as combustion of coals with trace uranium, windblown crustal material, and mining and processing of ores, whether it be for the uranium itself or other minerals such as rare earths and phosphate (Hamilton, 1970; Tadmor, 1986; Yunoki et al., 1995).

Natural uranium contains about 99.3% ^{238}U (half-life 4.468×10^9 yr), 0.7% ^{235}U (half-life 7.04×10^8 yr), and 0.0057% ^{234}U (half-life 2.46×10^5 yr). Aerosol particles containing uranium enriched in ^{235}U are definitely not from a natural source. Releases of enriched uranium and other radionuclides into the atmosphere have been reviewed by Salbu and Lind (2011). Besides major single releases such as the Chernobyl reactor failure or the burnup of the Cosmos satellite reactors (Krey et al., 1979; Leifer et al., 1987), longer-term sources of uranium such as windblown dust from nuclear test sites have been postulated to increase the ^{235}U content of atmospheric uranium (Kikiwada et al., 2009). Apart from isolated events, most of the uranium in the remote atmosphere is not enriched. Even during nuclear testing in the early 1960s, fallout increased $^{235}U/^{238}U$ in alpine ice from about

0.7% to about 0.8% (Warneke et al., 2002), so enriched uranium was a small fraction of total uranium.

Measurements of uranium in the atmosphere have generally been made with bulk filter samples that can process a large amount of air for detection of very low concentrations. Measurements of single particles can add important context: the matrix that the uranium is found in can constrain the sources. For example, (Utsonomiya et al., 2004) found uranium in particles from coal combustion. Other measurements of particles known to contain uranium have been made in the laboratory using thermal ionization (Stoffels and Lagergren, 1981; Kraiem et al., 2011), laser-induced breakdown spectroscopy (Fichet et al., 1999), laser ablation (Gieray et al., 1998), and secondary ion mass spectrometry (Ranebo et al., 2009).

In contrast, the particles described here were sampled by a mass spectrometer that flies on a research airplane to measure the composition of particles with no sample preparation. No special attempt was made to sample radiogenic or fissile material during atmospheric measurement campaigns performed with the spectrometer. Of particular interest is a particle apparently containing enriched uranium that was sampled by chance during one campaign at an altitude of about 7 km near the Aleutian Islands.

The Particle Analysis by Laser Mass Spectrometry (PALMS)

instrument samples the composition of single particles in the atmosphere with diameters between about 200 nm and 5 μm. Particles are brought into a vacuum and, when detected, hit with a pulse from a 193-nm laser as they travel through the instrument (Fig. S1). A time-of-flight mass spectrometer collects the entire mass spectrum of ions produced by the laser ablation. The mass spectrometer can record single spectra with high dynamic range, which aids in seeing isotopes of even minor species in the particles. PALMS also measures the diameter of each sampled particle, specifically the vacuum aerodynamic diameter which for spheres is the product of the geometric diameter and the density relative to 1.0 g cm^{-3}. The density enters to the first power rather than the square root often associated with aerodynamic diameters because the particle was smaller than the mean free path of air at the pressure in the portion of the PALMS vacuum inlet where the particle velocity is determined.

The PALMS instrument was originally constructed for high-altitude sampling (Thomson et al., 2000; Murphy et al., 2014) and has since been improved and converted for other research aircraft. Over the course of about 20 years PALMS has measured the composition of millions of particles in a wide variety of environments from 200 m above polar ice to 19 km in the tropical stratosphere. In August 2016 PALMS was sampling on the NASA DC-8 aircraft as part of the Atmospheric Tomography (ATom) program (https://espo.nasa.gov/missions/atom/content/ATom). The purpose of the field campaign was to obtain some of the first global cross-sections of the concentration of trace gases and of dust, smoke, and other particles in the remote troposphere over the Pacific and Atlantic Oceans.

1.1. Data

The particle under discussion was sampled on a research flight from Anchorage, Alaska to Kona, Hawaii at an altitude of about 7000 m above the Aleutian Islands on 3 August 2016 at 20:40 UTC. Fig. 1 presents the mass spectrum of the particle. Laser ablation of particles produces ions that are mostly either elements or fragments of the original molecules and rarely, parent peaks for the molecules. From the peak areas in the figure, the uranium ions are over 2% of the total ions produced by the laser ablation of this particle, one of the largest U signals seen in the history of the instrument. Most significantly, ^{235}U is 3.1 ± 0.5% of ^{238}U. This corresponds to low-enriched uranium typically used in nuclear reactors, whereas natural uranium contains only about 0.7% ^{235}U and weapons-grade uranium is enriched up to around 90% ^{235}U or more.

One must be confident that the m/z 235 peak is from ^{235}U. The strongest evidence is that, within uncertainty in the peak areas, the isotope ratio is the same for the doubly charged ions at m/z 117.5 and 119 as for m/z 235 and 238. This shows that m/z 235 is not due to a molecular ion; in the PALMS instrument doubly charged peaks are generally observed only for atomic ions. Although implausible, one must consider the possibility that some unusual spatial or energy separation during the laser ablation produced a "shoulder" on the 238 peak that happened to be both separated from the main peak and occur at m/z 235 rather than any other place. There are several lines of evidence against this. First, in Fig. 1 no other ion has such a shoulder. Second, three mass units would be a very large separation in a mass spectrometer that is compensated for initial ion energy and position. For example, one could hypothesize that the m/z 235 peak is due to ^{238}U ions that originated from a fragment of the particle that ended up in a different spatial position in the ion source than the main peak. But creating ions that arrive at the detector three mass units before their nominal arrival time would require a spatial separation that is not only many times larger than the diameter of the particle but also outside the ionization laser beam. Finally, the ratio of m/z 117.5 to 119 helps exclude the possibility that the m/z 235 peak is an artifact of the ionization mechanism. The production of doubly charged ions is very sensitive to ionization conditions, so any unusual ionization mechanism would be unlikely to produce the same amounts of singly and doubly charged ions. Singly and doubly charged uranium ions also have different loss rates as they interact with ablated neutral molecules (Gieray et al., 1998). Overall, we do not know of a plausible way to produce the observed spectrum unless the m/z 235 and m/z 117.5 peaks are from $^{235}U^+$ and $^{235}U^{++}$. The implausibility of creating an anomalous 235 peak in the ion source must be balanced against the rarity of ^{235}U.

The uncertainty in the isotope ratio is determined from the consistency of the singly and doubly charged ion ratios (about 0.2%) and the accuracy of PALMS for known isotopic ratios of similar size from other elements such as K or Fe (often about 10% of the smaller isotope). The UO^+ and UO_2^+ peaks show that the uranium was present as an oxide. The uranium peaks are broader than low-mass peaks in the spectrum such as C^+ and V^+. One reason is that uranium oxide is refractory. The ions formed from vaporized uranium oxide have more thermal energy, which broadens the peaks.

The largest peak in the mass spectrum is vanadium, and other major peaks include K^+, Fe^+, NO^+, and SO^+. The latter two peaks are most likely from nitrate and sulfate, which often condense on particles during atmospheric transport. There are a number of smaller peaks containing carbon. Vanadium in fine particles is considered a tracer for heavy oil combustion (Zoller et al., 1973; Viana et al., 2009). Except for the uranium, we have observed spectra similar to Fig. 1 in many particles, often at low altitude over the ocean near shipping lanes. Without
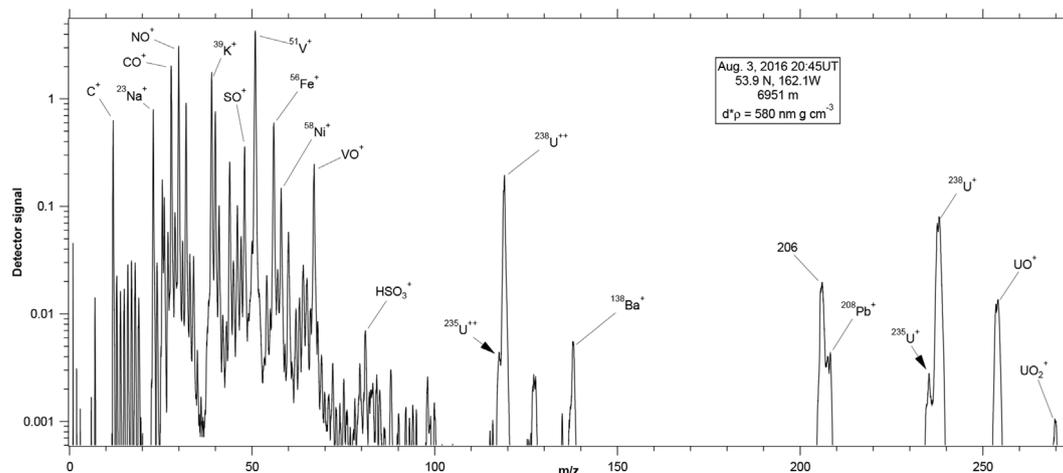

Fig. 1. Mass spectrum of the particle containing enriched uranium. The NO^+ peak indicates nitrates, SO^+ and HSO_3^+ sulfates, and abundant vanadium is usually a marker for heavy fuel oil. Many of the small, unmarked peaks at m/z < 100 are small fragments of organic molecules.
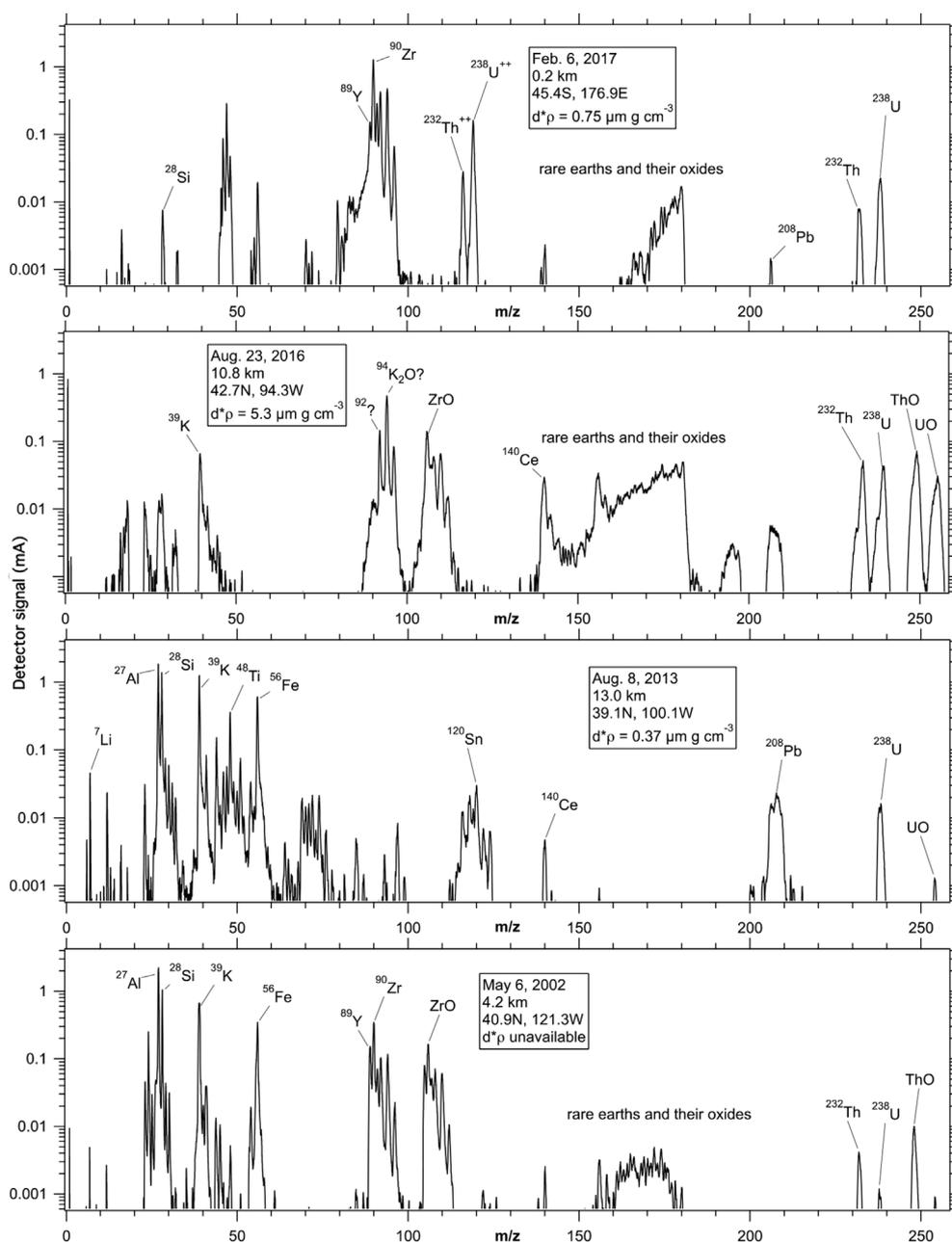
Fig. 2. Representative mass spectra of other particles containing uranium sampled with PALMS in other locations. These particles are consistent with uranium ores or other crustal material. All other uranium-containing particles sampled with PALMS through the years are consistent with ores, minerals, or similar material.

the uranium, we would have identified this particle as probable ship exhaust.

Metals are easily ionized in laser ablation of particles. Most of the mass of the particle was organic material, sulfates, and nitrates. The vanadium, uranium, and other metal peaks are large in Fig. 1 because of high sensitivity. We have not calibrated for uranium, but we have for several other metals, to which PALMS is quite sensitive. For example, 0.1% by mass Pb in ammonium nitrate produces about 3% Pb ions (Murphy et al., 2007). If U ionizes like Pb, then the mass fraction of U in the particle described here would be ~0.1 to 1%.

The vacuum aerodynamic diameter of the particle, which is the product of geometric diameter and density normalized to 1 g cm^{-3}, was 580 nm. The density of most sulfates and nitrates is less than 2 g cm^{-3}; most metal oxides are denser. If the average density were 2 g cm^{-3}, then the geometric diameter would be 290 nm. A lower average density would imply a larger geometric diameter and vice versa. Given that most of the particle was organics, nitrate and sulfates, the average density was probably ≤2 g cm^{-3}. If so, a 0.1 to 1% by mass U content

would represent 33 to 330 ag of U.

The association of uranium with vanadium is unique in the mass spectra we have recorded. Uranium is not especially common in submicron atmospheric particles (< 1 in 10000 particles have detectable uranium) but we have measured a number of particles with significant uranium content. All particles other than the one described here had compositions consistent with natural uranium-bearing minerals such as monazite. Examples are shown in Fig. 2. The peak in Fig. 1 at m/z 206 is also unusual, with no definite identification. If it is ^{206}Pb, the isotope ratio would be highly unusual compared to ^{208}Pb.

1.2. Possible regions of particle origin

A comprehensive suite of gas phase molecules was also measured on the aircraft. At the time the particle was sampled, the aircraft was conducting a vertical profile. The concentrations of ozone and carbon monoxide (about 76 and 97 ppbv respectively) were consistent with diluted pollution. Otherwise the air was not notable for unusual

Footprint emission sensitivity on 03 Aug 2016 20:39 (6951 m)
Maximum footprint value is 636.88 ×10^{-6} ns m^{-3}

0 1.5 3 6 12 24 47 94 188 375 750 1500
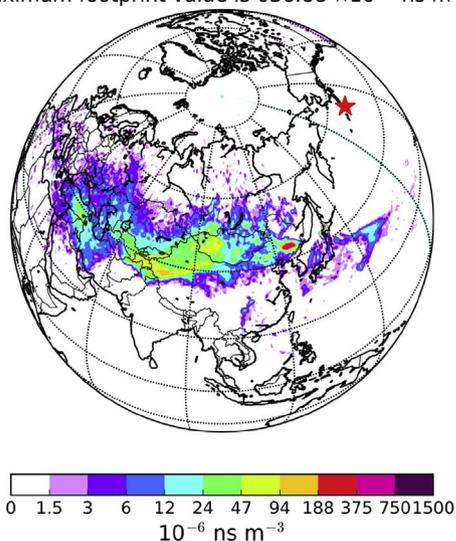10^{-6} ns m^{-3}

Fig. 3. Surface emission sensitivity footprint for the time and location of the sampled particle, indicated by the red star, from a FLEXPART 30-day retroplume calculation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

chemical gas phase chemistry. Two gas chromatograph systems measured a wide range of hydrocarbons and halocarbons. These samples also indicated enhanced pollution tracers, mostly as longer-lived anthropogenic molecules such as propane, benzene, and CH_2Cl_2 (with tropospheric lifetimes ranging from 2 weeks to several months). Shorter-lived molecules such as toluene (lifetime ≈ 3 days) were not enhanced. This is consistent with several days of transport over the Pacific Ocean. No unique signatures stood out.

Another possible clue to the origin of the air is the composition of particles measured nearby. Thallium was present in two particles measured within a minute (about 200 m in altitude) of the one containing uranium. In widespread sampling PALMS has detected thallium in very roughly 0.01% of particles, so finding two in 1 min (in this case about 120 particles) is extremely unusual. Several particles in this interval also contained Pb, Sn, or Sb. Sources of airborne thallium include Pb-Zn smelting and coal combustion (Belzile and Chen, 2017). A mass spectrum of one Tl-containing particle is included as Fig. S2. With S and Pb in addition to Tl but little or no Fe or alkali metals, it is more consistent with smelting than coal combustion.

To investigate the possible origin of the observed particle, we used two back trajectory models. The first was version 10 of the Lagrangian particle dispersion model FLEXPART (FLEXible PARTicle dispersion model, Stohl et al., 2005). The model was driven with 3-hourly operational meteorological analyses from the European Center for Medium-Range Weather Forecasts (ECMWF) with 137 vertical layers and a horizontal resolution of 0.5° × 0.5°. Computational trajectories were initiated at the location where the U-containing particle was sampled and were tracked 30 days back in time in FLEXPART's "retroplume" mode, which is a sufficient time to include most aerosol emissions arriving at the sampling location, given a typical aerosol lifetime (about 1 week). The tracking included gravitational settling characteristic for a spherical particle of the size observed and dry and wet deposition of gases or aerosols (Grythe et al., 2017). The simulation also includes turbulence (Cassiani et al., 2015), unresolved mesoscale motions (Stohl et al., 2005), and deep convection (Forster et al., 2007). Sensitivity calculations were performed for slightly different particle sizes and observation locations. The model output consists of an emission sensitivity, which is a quantitative measure for the particle mass concentration at the sampling location that would result from a unit emission flux at the Earth's surface. Since only a single particle was

observed, the emission sensitivity can also be interpreted as a probability distribution field of the particle's origin. We used it here to identify possible source regions of the particle.

Fig. 3 shows the results of the retroplume calculations. According to FLEXPART, the particle could have originated from a broad swath across Asia and beyond. Regions with higher probabilities than most are near the border between China and North Korea, the Tarim basin in China, or west of Hokkaido Island in Japan. Sensitivity calculations with particles of different size and density (thus, different gravitational settling speeds) as well as using starting positions 150 m below and 120 m above the actual observation location resulted in similar retroplumes, indicating that the results are relatively robust. Nevertheless, one has to consider that transport calculations over such long distances are always quite sensitive to even small errors in the meteorological analyses used. Therefore, other source regions cannot be excluded with certainty.

Fig. 4 shows the results from the second back trajectory model, which uses the Bowman trajectory code (Bowman and Carrie, 2002; Bowman et al., 2013) with NCEP (National Centers for Environmental Prediction) meteorological fields. For this analysis, a cluster of trajectories was initialized every minute centered along the flight track. The spread in the cluster (light green lines) gives an indication of the uncertainty in the trajectory released from the exact flight track location (orange line). In the bottom plot of Fig. 4, the location where any of the cluster of trajectories entered the boundary layer is indicated by a blue shaded region on the map. The shading indicates the normalized probability that any trajectory at any time within the 10 days prior to the flight time entered the boundary layer in a specific region. The trajectories show similar features to the FLEXPART retroplumes. Both models show a transition along the flight track from outflow from Asia to marine air above that, in agreement with the aircraft observations that show CO dropping by about 30 ppbv just after (above) the uranium particle was sampled. The models, however, put the transition about 500 m higher than observed. The Bowman NCEP analysis puts the trajectories over Asia slightly south of the FLEXPART model. Nevertheless, the two models are in general agreement about the direction of possible source locations.

2. Discussion

We have sampled numerous other particles with trace uranium, almost always with lower content than this particle. All of the other particles have been consistent with natural uranium. In those particles, not only was the isotope ratio not enriched, but the uranium was always present with rare earths or other elements consistent with uranium minerals and ores.

The data do not show any obvious source for this particle. The isotope ratio indicates relatively fresh reactor grade uranium. One possible source is that it escaped during production of reactor fuel, which is done on an industrial scale. This, however, does not explain the association with oil combustion products. In addition, the amount of uranium in the particle is equivalent to a sphere less than 60 nm in diameter. Mechanical processes producing uranium dust would generally produce much larger particles, such as the uranium particles from a nuclear facility that were analyzed by Kraiem et al. (2011). Instead, sub-100-nm particles are often produced from condensation of vapor from high temperature processes (Taylor and Flagan, 1981). For example, uranium-rich inclusions of about 10 nm have been found in particulate matter from coal combustion (Utsonomiya et al., 2004).

Another possible source is resuspension of a particle from a prior nuclear accident, such as Chernobyl or Fukushima. The lifetime of aerosol particles in the troposphere is far too short for a particle have stayed in the atmosphere since the accidents. In the days prior to sampling, there were no major fires or other known events near Chernobyl or Fukushima to resuspend particles. A wildfire would not explain the association with heavy oil combustion products. In
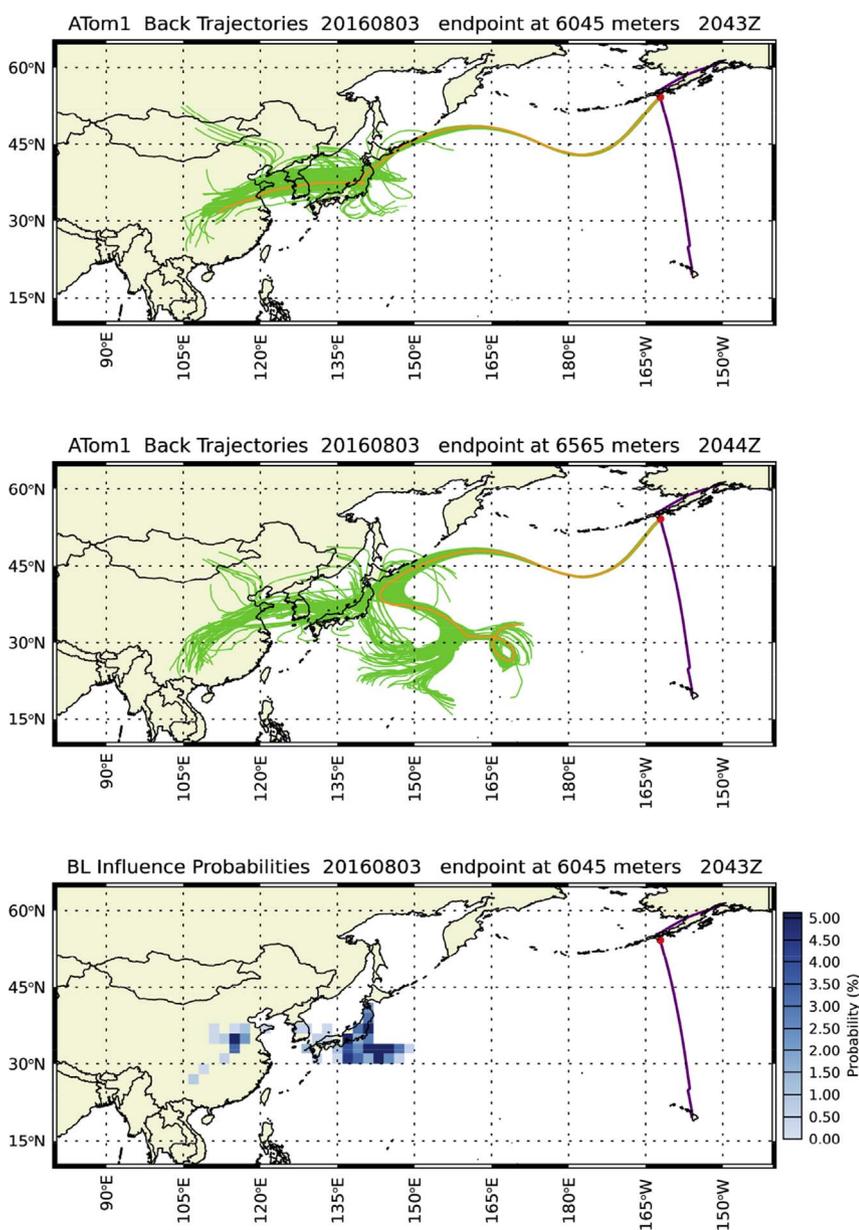
Fig. 4. 10-day back trajectories based on NCEP meteorology initialized on the aircraft track just before the particle containing enriched uranium was sampled (top and middle plots). The light green lines show individual trajectories initialized from a cluster centered on a single location on the flight track. The orange line is the trajectory from the center of the cluster at the exact flight track location. The purple line is the flight track and the red symbol is the location on the track where the trajectories were initialized. The bottom plot shows the locations where trajectories entered the boundary layer at any point in the previous 10 days shaded blue by the normalized probability of boundary layer influence. Times just prior to sampling are chosen because gas-phase molecules show the particle was sampled before the actual transition between Asian and marine air. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

addition, in the particle there are no other elements associated with reactors above the detection limit, such as fission products or zirconium from fuel cladding.

It is unlikely, but possible, that the oil combustion products were in a separate particle that coagulated with a uranium particle in the atmosphere. The most common particles available for a nanoparticle to coagulate with are organic-sulfate mixtures, not oil combustion particles. Even if there were coagulation, the observed particle would still require the production and dispersion into the air of an enriched uranium oxide particle in a size range usually associated with combustion processes.

It is not possible to estimate an emission amount from sampling one particle. We can say that PALMS is very sensitive to dispersed trace metals. One comparison is to another heavy metal, Pb. Over the United States, PALMS has sampled thousands of particles containing Pb from the ~500 tons per year of Pb in leaded fuel used in light piston aircraft (Murphy et al., 2007). A rough corollary is that for a small amount of metal dispersed as fine particles in a continent-sized amount of air, the odds of sampling one particle would be very small, but not astronomically against sampling one.

In summary, the data would suggest that either a tiny (< 60 nm) particle of nearly pure enriched uranium oxide coagulated with an oil combustion particle, or the particle originated from combustion of heavy oil contaminated with enriched uranium. Neither explanation is especially satisfactory. The particle was probably emitted within a broad region that includes Japan, China, and South and North Korea. This particle could suggest a novel process or event whereby enriched uranium was released into the atmosphere.

Conflicts of interest

None.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2018.01.006.

References

Belzile, N., Chen, Y.-W., 2017. Thallium in the environment: a critical review focused on natural waters, soils, sediments and airborne particles. Appl. Geochem. 84, 218–243.

Bowman, K.P., Carrie, G.D., 2002. The mean-meridional transport circulation of the troposphere in an idealized GCM. J. Atmos. Sci. 59, 1502–1514.

Bowman, K.P., Lin, J.C., Stohl, A., Draxler, R., Konopka, P., Andrews, A., Brunner, D., 2013. Input data requirements Lagrangian trajectory models. Bull. Am. Meteorol. Soc. 94, 1051–1058. http://dx.doi.org/10.1175/BAMS-D-12-00076.1.

Cassiani, M., Stohl, A., Brioude, J., 2015. Lagrangian stochastic modelling of dispersion in the convective boundary layer with skewed turbulence conditions and a vertical density gradient: formulation and implementation in the 30 FLEXPART model. Boundary-Layer Meteorol. 154, 367–390.

Fichet, P., Mauchien, P., Moulin, C., 1999. Determination of impurities in uranium and plutonium dioxides by laser-induced breakdown spectroscopy. Appl. Spectrosc. 53, 1111–1117.

Forster, C., Stohl, A., Seibert, P., 2007. Parameterization of convective transport in a Lagrangian particle dispersion model and its evaluation. J. Appl. Met. Clim. 46, 403–422.

Gieray, R.A., Reilly, P.T.A., Yang, M., Whitten, W.B., Ramsey, J.M., 1998. Tandem mass spectrometry of uranium and uranium oxides in airborne particulates. Anal. Chem. 70, 117–120 1998.

Grythe, H., Kristiansen, N.I., Groot Zwaaftink, C.D., Eckhardt, S., Ström, J., Tunved, P., Krejci, R., Stohl, A., 2017. A new aerosol wet removal scheme for the Lagrangian particle model FLEXPART. Geosci. Model Dev. Discuss. 10 1447–1446.

Hamilton, E.I., 1970. The concentration of uranium in air from contrasted natural environments. Health Phys. 19, 511–520.

Kikiwada, Y., Oda, K., Yamauchi, R., Nomura, M., Honda, T., Io, T., Hirose, K., Igarashi, Y., 2009. Anomalous uranium isotope ratio in atmospheric deposits in Japan. J. Nucl. Sci. Technol. 46, 1094–1098.

Kraiem, M., Richter, S., Kühn, H., Stefaniak, E.A., Kerckhove, G., Truyens, J., Aregbe, Y., 2011. Investigation of uranium isotopic signatures in real-life particles from a nuclear facility by thermal ionization mass spectrometry. Anal. Chem. 83, 3011–3016 2011.

Krey, P.W., Leifer, R., Benson, W.K., Dietz, L.A., Hendrikson, H.C., Colluzza, J.L., 1979. Atmospheric burnup of the Cosmos-954 reactor. Science 205, 583–585.

Leifer, R., Juzdan, R.Z., Kelly, W.R., Fassett, J.D., Eberhardt, K.R., 1987. Detection of uranium from Cosmos-1402 in the stratosphere. Science 238, 512–514.

Murphy, D.M., Hudson, P.K., Cziczo, D.J., Gallavardin, S., Froyd, K.D., Johnston, M.V., Middlebrook, A.M., Reinard, M.S., Thomson, D.S., Thornberry, T., Wexler, A.S., 2007. Distribution of lead in single atmospheric particles. Atmos. Chem. Phys. 7, 3195–3210.

Murphy, D.M., Froyd, K.D., Schwarz, J.P., Wilson, J.C., 2014. The chemical composition of stratospheric aerosol particles. Q. J. R. Meteorol. Soc. 140, 1269–1278. http://dx.doi.org/10.1002/qj.2213.

Ranebo, Y., Hedberg, P.M.L., Whitehouse, M.J., Ingenerid, K., Littmann, S., 2009. Improved isotopic SIMS measurements of uranium particles for nuclear safeguard purposes. J. Anal. Atomic Spectrom. 24, 277–287.

Salbu, B., Lind, O.C., 2011. Radioactive particles released into the environment from nuclear events. In: Kalmykov, S., Denecke, M. (Eds.), Actinide Nanoparticle Research. Springer, Berlin, Heidelberg.

Stoffels, J.J., Lagergren, C.R., 1981. On the real-time measurement of particles in air by direct-inlet surface-ionization mass spectrometry. Int. J. Mass Spectrom. Ion Phys. 40, 243–254.

Stohl, A., Forster, C., Frank, A., Seibert, P., Wotawa, G., 2005. Technical Note: The Lagrangian particle dispersion model FLEXPART version 6.2. Atmos. Chem. Phys. 5, 2461–2474. http://dx.doi.org/10.5194/acp-5-2461-2005.

Tadmor, J., 1986. Radioactivity from coal-fired power plants: a review. J. Environ. Radioact. 4, 177–204.

Taylor, D.D., Flagan, R.C., 1981. The influence of combustor operation on fine particles from coal combustion. Aerosol Sci. Technol. 1, 103–117.

Thomson, D.S., Schein, M.E., Murphy, D.M., 2000. Particle analysis by laser mass spectrometry WB-57F instrument overview. Aero. Sci. Technol. 33, 153–169.

Utsonomiya, S., Jensen, K.A., Keeler, G.J., Ewing, R.C., 2004. Uraninite and fullerene in atmospheric particulates. Environ. Sci. Technol. 36, 4923–4947.

Viana, M., Amato, F., Alastuey, A., Querol, X., Moreno, T., Garcia, S., Santos, D., Herce, M.D., Fernandez-Patier, R., 2009. Chemical tracers of particulate emissions from commercial shipping. Environ. Sci. Technol. 43, 7472–7477.

Warneke, T., Croudace, I.W., Warwick, P.E., Taylor, R.N., 2002. A new ground-level fallout record of uranium and plutonium isotopes for northern temperate latitudes. Earth Planet Sci. Lett. 2003, 1047–1057.

Yunoki, E., Kataoka, T., Michihiro, K., Sugiyama, H., Shimizu, M., Mori, T., 1995. Background levels of ^{238}U and ^{226}Ra in atmospheric aerosols. J. Radioanal. Nucl. Chem. 189, 157–164.

Zoller, W.H., Gordon, G.E., Gladney, E.S., Jones, A.G., 1973. The sources and distribution of vanadium in the atmosphere. In: "Trace Elements in the Environment", Advances in Chemistry, vol. 123 American Chemical Society, Washington, DC ISBN13: 9780841201859.