Air Fluorescence Method for Remote Measurement of Alpha Surface Contamination: Research Progress and Prospects

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Abstract

In the nuclear industry, numerous facilities and plants exhibit α surface contamination. During emergency scenarios, remote detection of anomalous areas is typically required first, followed by the scientific development of remediation strategies based on measurement results. To address this need, an α particle measurement technique based on air fluorescence has been proposed internationally. The principle relies on nitrogen in air undergoing excitation-deexcitation processes under α radiation, emitting ultraviolet fluorescence at specific wavelengths that can be remotely detected by UV cameras, thus enabling indirect detection of α contamination. Research reports on α contamination remote detection technology are relatively scarce domestically; CNNC 404, in collaboration with Lanzhou University, has pioneered investigations in this area. This paper primarily reviews the international development status of α contamination remote detection technology based on nitrogen fluorescence, outlines the latest research efforts of our team, and aims to provide a reference for domestic research in α contamination remote detection.

Full Text

Preamble

Research Progress and Prospects of Remote α Surface Contamination Measurement Devices Using Air Fluorescence Method

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Abstract

In the nuclear industry, α surface contamination exists in numerous facilities and sites. During emergency situations, remote detection of contaminated areas is typically required before scientifically formulating response plans based on measurement results. To address this need, international researchers have proposed an α particle measurement method based on air fluorescence. The principle is that nitrogen molecules in air, when excited by α radiation, undergo de-excitation processes that emit ultraviolet fluorescence at specific wavelengths, which can be remotely detected by UV cameras to indirectly identify α contamination. Domestic research on remote α contamination detection technology remains limited, with China National Nuclear Corporation 404 Company Limited and Lanzhou University pioneering investigations in this area. This paper reviews international developments in remote α contamination detection technology based on nitrogen fluorescence, summarizes the current team' s latest research efforts, and aims to provide references for advancing domestic research on remote α contamination detection.

Keywords: Air fluorescence; α surface contamination; Remote detection; Nuclear detection

1. Principle of α Contamination Imaging

α rays are charged particle streams that cause ionization and excitation of nitrogen molecules in air, releasing energy in the form of photons within the ultraviolet range [**?**, **?**]. These photons have much longer mean free paths than α particles themselves, enabling remote detection through α-induced radioluminescence in air. Research on air fluorescence dates back to the work of the Huggins couple in 1903 [**?**], but it was not until 2001 that this effect began to be applied for remote detection of α radioactivity [**?**, **?**]. Ukrainian scientist Baschenko conducted spectral studies in 2004, obtaining the radioluminescence spectrum of α radiation in standard atmospheric conditions, as shown in [Figure 1: see original paper] [**?**]. Over 95% of α-induced radioluminescence intensity concentrates in the 300–400 nm wavelength range, corresponding to the near-ultraviolet region (UVA and UVB), with nitrogen molecules being the primary emitters.

[Figure 1: see original paper]

The radioluminescence effect is applicable to optical measurement of all ionizing radiation but is particularly suitable for α radiation. Due to α rays' short range and high ionization density, their energy deposits intensively within a few centimeters of the source, facilitating source localization [**?**, **?**]. Baschenko also found that the intensity ratio of radioluminescence induced by α, β, and γ radiation is $1{:}10^{-8}{:}10^{-10}$, allowing α radiation to be distinguished even in the presence of other radiation types due to its significantly stronger optical signal [**?**].

However, the 300–400 nm wavelength range of α-induced fluorescence overlaps with solar spectra and typical fluorescent lighting, creating severe background interference [**?**]. Solar radiation intensity in this band reaches (2–8)$×10^{-2}$ $\mathrm{W \cdot cm^{-2} \cdot nm^{-1}}$, while the peak radioluminescence intensity from a 37 MBq source ranges between 10^{-10}–10^{-7} $\mathrm{W \cdot cm^{-2} \cdot nm^{-1}}$. Even at night, ambient light exceeds the radioluminescence signal [**?**, **?**]. To minimize interference, reported α contamination imaging devices typically operate in dark environments or under special lighting conditions to reduce background pressure [**?**, **?**, **?**, **?**, **?**]. Another strategy targets the ultraviolet C band (UVC, 180–280 nm), known as the "solar blind region" [**?**]. Although fluorescence intensity is lower in this band, solar radiation is absorbed by atmospheric ozone and cannot reach the surface, while window glass effectively suppresses deep UV radiation from common light sources, resulting in extremely low background interference [**?**]. Research on remote α contamination imaging in the solar blind region under normal lighting conditions has also progressed [**?**, **?**].

2. Technical Selection for α Contamination Remote Imaging Systems

A remote α contamination imaging device generally consists of an optical system and a photoelectric signal conversion system, with data processed by a computer to generate contamination images. Auxiliary components can be added to improve imaging performance, as illustrated in [Figure 2: see original paper].

[Figure 2: see original paper]

The following sections outline the technical choices for each component of α contamination remote imaging systems.

2.1 Optical System

The optical system efficiently collects fluorescence and transmits it to subsequent imaging equipment. Literature reports indicate that most systems employ fused silica or quartz glass optical components (which transmit UV light) for objectives, focusing lenses, and filters [**?**, **?**, **?**, **?**]. For example, Inrig et al. designed a detector optical system comprising six spherical lenses and one aspherical lens (phase mask), all made of fused silica with anti-reflection coatings to reduce light loss. This design sacrificed some field of view and resolution (with an f-number

of only 1.23) to increase light throughput [?]. Johan Sand et al. designed an optical system with stacked filters that maximally attenuates visible light while maintaining high UV transmission, enabling detection under normal lighting conditions [?].

A significant challenge in nitrogen fluorescence-based α contamination detection is background light interference (natural or artificial lighting), which typically far exceeds the intensity of α-induced nitrogen fluorescence signals [?]. While operating in dark or special lighting conditions can alleviate this issue, practical applications cannot always dictate the illumination at detection sites. Therefore, optical systems must incorporate optimized filtering to reduce background interference.

[Figure 3: see original paper]

2.2 Photoelectric Signal Conversion System

Photodetectors convert optical signals into electrical signals. To date, reported α contamination imaging systems have exclusively used photomultiplier tubes (PMTs) or charge-coupled devices (CCDs). Although Shaw et al. reported a Geiger-mode avalanche photodiode (GM-APD) in 2019 with better quantum efficiency than CCDs and PMTs for deep UV detection, no applications to remote α detection have been published [?].

As early as 2004, Ukrainian scientist Baschenko used a PMT as the photodetector in a laboratory model measuring the atmospheric α radioluminescence spectrum under standard conditions [?]. PMTs offer good single-photon sensitivity and low dark count rates, though they lack imaging capability, they are cost-effective [?]. Hamamatsu Corporation has developed a cesium-telluride photocathode PMT (H11870-09) with UV response from 160–320 nm, suitable for far-UV detection and known as a solar-blind (SB) detector [?]. While optical filtering reduces background light, it also attenuates the already weak fluorescence signal. An alternative strategy employs photodetectors insensitive to visible light but sensitive to the solar blind region. Building upon filtering, Johan Sand et al. used this cesium-telluride photocathode PMT to develop an α contamination detector operable in daylight, achieving high detection efficiency at moderate cost [?]. Another widely reported detector, UVTron (R9533, Hamamatsu), features an even narrower far-UV response range (180–260 nm) that better matches the solar blind region [?]. Crompton et al. employed this UVTron detector in their α contamination imaging system, enabling operation under normal lighting conditions [?].

CCD detectors, unlike PMTs, offer imaging capability without requiring point-by-point scanning and are widely used in scientific cameras. Researchers led by Lamadie pioneered remote α contamination imaging devices based on air fluorescence technology, using CCD cameras to detect radioluminescence signals with liquid nitrogen cooling to reduce noise and enable longer exposures for more accurate images [?]. As CCD technology advanced, intensified CCDs (ICCDs)

and electron-multiplying CCDs (EMCCDs) were developed for extremely low-light imaging. In 2015, Johan Sand et al. compared the performance of ICCD- and EMCCD-based α contamination detectors, both yielding good results [**?**]. However, Lamadie' s and Sand' s studies were conducted in dark conditions, and CCD cameras require special treatments like liquid nitrogen cooling. Daylight operation with CCD-based α contamination imaging demands superior filtering. Ivanov et al. successfully used a commercial DayCor SuperB UV camera for daylight remote α contamination imaging. Originally designed for corona and arc detection in high-voltage equipment, this camera combines an ICCCD with optical filtering to detect only UV radiation below 290 nm (UVC) [**?**]. Nevertheless, Lamadie and Sand suggested focusing on improvements in optical filtering and photodetection materials, developing new materials such as gallium nitride (GaN) that might replace CCDs in commercial applications [**?**, **?**]. Additionally, ICCDs and EMCCDs, as scientific components with military applications, are subject to export controls.

2.3 Auxiliary Systems

Beyond the optical and photoelectric conversion systems, auxiliary methods can enhance imaging performance or expand system functionality. For instance, nitrogen fluorescence in atmospheric environments can be effectively quenched by oxygen and water vapor, but fluorescence yield increases significantly in pure nitrogen or inert gas environments [**?**, **?**]. Crompton et al. used a gas delivery system to flow inert gases over radiation sources, investigating enhancement effects of Ar, Xe, Ne, N_2, and Kr gases for detecting weaker α sources [**?**]. Johan Sand' s team developed a UV-γ coincidence imaging system ([Figure 4: see original paper]) that adds a γ detector and gating mechanism to an α contamination detector, recording only γ photons detected simultaneously with radioluminescence to identify α radiation in high-radiation backgrounds [**?**]. In Kume et al.' s study, a specially structured lead box was configured around the photodetector to remove γ-ray background, with simulations of γ-ray attenuation used to optimize the lead box structure for better shielding [**?**].

While advances in optical filtering and photodetector technology may eventually eliminate the need for these auxiliary methods, they currently provide positive contributions to improving signal-to-noise ratios and lowering detection limits.

[Figure 4: see original paper]

3. Several Promising α Contamination Detection Devices and Their Performance

Before evaluating α contamination remote detectors, it is necessary to understand practical detection requirements. Kume et al. noted that radioactive waste from nuclear power plants exceeding 3.7 MBq/kg is classified as transuranic (TRU) waste and must be separated from other waste. Therefore, the standard

for α detectors in nuclear power plant operations is the ability to resolve α radiation levels of 3.7 MBq/kg or lower in high-radiation environments [**?**]. However, actual conditions are more complex, with α contamination intensities in nuclear facilities potentially ranging from Bq to GBq levels [**?**]. This section chronologically introduces several high-performance α contamination remote detectors with detailed parameters.

In 2005, Lamadie et al. developed a fluorescence-based remote α contamination imaging device consisting of a radiation-collecting objective lens and a CCD detector. The system could detect point and extended sources at levels as low as 430 Bq/cm^2 at a 20 cm test distance, and could detect through translucent materials like 10 mm Plexiglas (with 600 s exposure time), but required dark conditions [**?**].

In 2009 and 2011, Russian scientists Ivanov et al. selected a DayCor SuperB UV camera for α contamination imaging. Test results showed a minimum detectable activity of 10^5 Bq for point sources at 3 m distance with 600 s exposure time, with sensitivity improving through longer CCD exposures [**?**, **?**]. Ivanov' s detector could operate in daylight conditions.

In 2011, Inrig et al. reported an α contamination camera incorporating laser alignment, ultrasonic ranging, conventional photography, and optical movement/rotation components. The camera could detect α sources with activity of approximately 1 Ci (0.037 MBq) at 1.5 m distance (exposure time not specified for this minimum detectable activity), with tests conducted in dark conditions [**?**]. Imaging results are shown in [Figure 5: see original paper].

[Figure 5: see original paper]

Johan Sand' s team reported their HAUVA remote α contamination detection device in 2010 [**?**]. The system used continuous filters to form an interference filter and beam splitter, dividing incident light into two paths to detect α-induced radioluminescence signals and background illumination separately, distinguishing weak fluorescence from background through spectral filtering. Building on this, they investigated a time-coincidence detection scheme that similarly split incident light into two paths to measure the temporal distribution of photons from single radiation decay events. Since fluorescence decay time from a single α event is 5 ns, background coincidence events on this timescale are extremely rare, enabling time-coincidence filtering to differentiate α-induced fluorescence from stronger background illumination. Experimental results demonstrated that HAUVA could detect a 100 kBq point source at 0.4 m within 1 s under bright yellow fluorescent or white LED lighting.

Recognizing the potential of time-coincidence filtering, Johan Sand' s team developed a fluorescence-based UV-γ coincidence imaging system in 2013, again applying time-coincidence principles (device composition described in Section 2.3). The γ detector only triggers when α-induced fluorescence photons are detected, ensuring the system focuses exclusively on α radiation signals. This

UV-γ coincidence technique enhances α detection in high-activity backgrounds [**?**].

In the same year, Kume et al. developed an α camera that detected a 1.5 kBq α source at 1 m distance within 30 s. While tests were conducted in darkness, the authors noted that future commercial versions could incorporate filters to enable field operation [**?**].

In 2015, Johan Sand' s team field-tested two UV cameras based on ICCD and EMCCD technologies for α contamination detection. Both could detect MBq-level α radiation within 100 s at 0.5 m distance, though tests were performed in dark conditions [**?**]. In 2016, the team developed another α contamination detector operable under strong lighting, using two PMTs sensitive to far-UV and near-UV light respectively, each matched with specific filter stacks. The detector for illuminated environments could detect α sources with minimum activity of 800 kBq within 10 s under bright fluorescent lighting. Sensitivity could be further improved using nitrogen or argon gas purging [**?**].

In 2017, Mahé and Venara from the French Alternative Energies and Atomic Energy Commission (CEA) Nuclear Measurement Laboratory reported their patented single-unit dual-camera detector for in-situ α/γ measurement, though specific performance parameters were not provided [**?**].

Crompton et al. reported their UVTron-based α contamination imaging device in 2017 and 2018 (operable under lighting conditions) and explored fluorescence enhancement under inert gas conditions. As shown in [Figure 6: see original paper], the detector was placed outside a gas flow box approximately 20 mm from the radiation source. Calculations indicated that the minimum detectable activity in air within 3600 s ranged between 89–149 kBq, while xenon flow enhancement could lower the detection limit to approximately 47 kBq [**?**].

[Figure 6: see original paper]

Recent remote α contamination detection research has increasingly focused on the solar blind region. In 2020, Gamage and Crompton et al. revisited UVTron sensor-based remote α contamination detection [**?**], this time placing the UVTron sensor in a tungsten collimator and using various radioisotopes (^{210}Po, ^{241}Am, ^{137}Cs, ^{90}Sr, and ^{60}Co) to investigate shielding effectiveness against γ and β radiation. Results showed that while the collimator somewhat protected the UVTron sensor from γ and β radiation, it also significantly reduced the UVC signal.

In 2021, Krasniqi and Kerst et al. developed a solar blind region-based remote α contamination detector operable under strong light. The detector featured two optical systems: an ICCD camera for visible light imaging and a cesium-telluride photocathode PMT with optics for α-induced radioluminescence imaging. Notably, Kerst et al. enhanced UVC band radioluminescence by flowing NO gas, enabling imaging of low-activity α sources, with results shown in [Figure 7: see original paper] [**?**]. The primary difficulty in solar blind region imaging is

the extremely low intensity of α-induced radioluminescence. In 2018, Kerst et al. studied nitrogen' s effect on UVC band α-induced radioluminescence, finding that adding trace NO to the N_2 atmosphere around samples increased overall radioluminescence yield by approximately 25-fold, with nearly all emission falling within the UVC band [**?**]. This discovery was successfully applied in their 2021 study, demonstrating significant potential for solar blind region radioluminescence imaging.

[Figure 7: see original paper]

4. Analysis and Summary

Evaluating an α contamination detector requires comprehensive consideration of detection distance, minimum detectable activity, measurement time, and lighting conditions. summarizes the performance parameters of the aforementioned α contamination remote detectors, with data extracted from literature.

In recent years, the joint team from China National Nuclear Corporation 404 Company Limited and Lanzhou University has conducted systematic research on key technologies for α particle air fluorescence measurement. By comparing data from existing literature, we established a simulation model program suitable for α-induced nitrogen fluorescence yield. CCD camera selection and research were performed, with dark current noise, readout noise, shot noise, and excess noise factor introduced into imaging simulations to thoroughly investigate critical parameters including imaging signal-to-noise ratio (SNR), resolution, and interference between strong and weak sources [**?**]. Based on relevant theories and thermal experimental requirements reported in literature, and supported by the CNNC "Young Talented" research project, the team constructed an α particle air fluorescence measurement and collection system ([Figure 8: see original paper]). The entire apparatus consists of three parts: a darkroom, vacuum chamber, and electronic acquisition system. By simulating different gas environments in the vacuum chamber, experimental measurements of α-induced gas fluorescence yield were conducted using a PMT and backend signal acquisition system. The α source was placed inside the chamber, which connects to the darkroom through a viewing window, allowing the PMT in the darkroom to measure fluorescence induced by the α source (details in Section 4.2). Based on this system and thermal experimental results, preliminary design concepts were developed for an air fluorescence-based remote α surface contamination measurement device.

[Figure 8: see original paper]

4.1 Simulation Work Based on Geant4 [**?**]

Building on previous research, we identified similarities in nitrogen fluorescence spectra excited by different charged particles, where spectral lines originate from specific molecular energy state transitions. Drawing from high-energy cosmic ray electron-induced nitrogen fluorescence studies, we borrowed the physical

mechanisms of nitrogen fluorescence generation and de-excitation to develop a physical model package for charged particle-induced nitrogen fluorescence using Geant4 tools. [Figure 9: see original paper] illustrates the physical processes of α particle interactions with gas molecules and the computational model geometry. The model package comprises five components:

[Figure 9: see original paper]

1. Incident particles (α particles, electrons, and gamma rays) interact with gas molecules in the medium. For α particle ionization, we adopted the effective charge-corrected Rudd model [**?**, **?**], while nuclear stopping used Geant4' s built-in physical model. Electron interactions with the medium, including elastic and inelastic collision cross-sections, referenced the extended Opal model [**?**], which extends the Opal model' s energy range to the keV scale. For lower-energy electron interactions, we relied on experimental cross-section data compiled by Itikawa [**?**, **?**, **?**]. Electron scattering angles were adapted from Geant4' s built-in ionization model, with isotropic treatment applied near very low electron energies. Gamma ray interactions (photoelectric effect, Compton effect, electron-positron pair production) directly used Geant4' s built-in processes.

2. Any primary particle generates secondary electrons through medium interactions, which continue interacting according to electron-medium interaction models. These secondary electrons interact with nitrogen molecules through elastic collisions, inelastic collisions, or ionization excitation. Only inelastic scattering and ionization excitation can induce nitrogen fluorescence. By comparing the total cross-section in the electron model [**?**] with non-ionizing excitation and ionization excitation cross-sections, we determined the probability of electrons exciting nitrogen molecules to the 2P system (non-ionizing excitation) or 1N system (ionization excitation). Since non-ionizing excitation cross-section becomes negligible above 100 eV electron energy, high-energy electrons (>120 eV) primarily produce fluorescence from the 1N system [**?**, **?**, **?**].

3. For nitrogen molecules excited to the 2P or 1N systems, Franck-Condon factors and Einstein coefficients [**?**] determine the probability of occupying these states and transitioning to other states within the system.

4. For the de-excitation process, molecular collision quenching probability is introduced, where quenching occurs without photon emission. The collision quenching probability follows the relation: P_{quench} = (P/(kT)) $\times$ Σ(f_x $\times$ Q_x), where P is gas medium pressure, k is Boltzmann' s constant, T is temperature in Kelvin, f_x is the volume fraction of gas component x, and Q_x is the quenching factor for gas component x.

5. Combining steps (3) and (4), the final de-excitation mode of nitrogen molecules in the 2P or 1N systems is determined, including whether photons are produced and their corresponding wavelengths. Simulation results from this toolkit show good agreement with existing experimental data

[?]. Based on this, we simulated collection efficiency variations for fluorescence at different distances and detector sizes, providing prior knowledge for subsequent experiments.

Additionally, referencing parameters of Andor' s Ixon Ultra 888 camera, we used this model package to simulate imaging SNR, resolution, and masking effects, with results shown in [Figure 10: see original paper]. The findings indicate: (1) For α sources at the same activity level, SNR gradually increases with imaging time; at constant imaging time, higher-activity α sources yield higher SNR. (2) With this camera' s parameters, the imaging resolution for α-induced fluorescence is remarkably high, with source boundaries nearly indistinguishable from actual source dimensions. (3) Adjacent strong sources exhibit a "masking" effect on nearby weak sources, which diminishes with increasing separation distance. These results provide important references for designing key engineering metrics including imaging SNR, imaging time, minimum detectable activity, CCD camera selection, and imaging resolution.

[Figure 10: see original paper]

4.2 Precise Experimental Measurement of Nitrogen Fluorescence Yield

First, collisional quenching between gas molecules causes α-induced nitrogen fluorescence yield to vary with environmental conditions, interfering with quantitative analysis in remote α radioactivity measurements. Therefore, precise measurement of α-induced nitrogen fluorescence yield under various conditions (temperature, humidity, pressure, gas flow rate, etc.) is necessary for subsequent data correction.

[Figure 11: see original paper] shows the experimental measurement schematic for α-induced nitrogen fluorescence yield corresponding to [Figure 8: see original paper]. The vacuum chamber provides a relatively sealed environment to simulate complex atmospheric conditions (different temperatures, humidity, pressures, and gas flow rates) on a small scale. Temperature control uses a "temperature sensor + heating tape + solenoid valve" approach, heating when temperature falls below the setpoint and stopping when exceeded. Humidity control relies on the dynamic equilibrium characteristic of saturated salt solutions in sealed spaces, using six different saturated salt solutions (lithium chloride, potassium acetate, lithium fluoride, sodium bromide, potassium iodide, and potassium bromide) to achieve different humidity gradients. Pressure control is achieved through vacuum chamber and valve regulation. Gas flow rate effects were verified experimentally outside the vacuum chamber. Internal temperature, humidity, and pressure sensors enable real-time readout of environmental parameters. The viewing window on one side of the vacuum chamber extends into the darkroom, allowing fluorescence induced by the α source inside to be detected by the PMT in the darkroom. Guided by Monte Carlo simulation results, we adopted a "filter + large-area PMT" scheme to simplify the optical

path, avoid unnecessary attenuation corrections, reduce background at other wavelengths, and ensure maximum fluorescence photon collection. Additionally, coincidence between the PMT and Si detector inside the vacuum chamber temporally constrains background and noise, further improving SNR for optimal experimental measurement results in weak fluorescence environments. The electronic system provides bias voltage for the Si detector, preamplifier power, and high voltage for the PMT. Signals from the PMT and Si detector are input to a multi-channel analyzer for event acquisition, as shown in [Figure 11: see original paper].

[Figure 11: see original paper]

Based on the theoretical expression [**?**] and experimental definition of α-induced nitrogen fluorescence yield:

Y_{exp} = N_{detected} / (E_α × t × Ω × × T_{filter})

where Y_{th} is the intrinsic fluorescence yield, Y_{exp} is the detected yield, P_{quench}^{intrinsic} is the intrinsic quenching probability, Ω is the solid angle at the focusing lens window, and is the surface viewing angle of the monitored material. A correction function software based on this relationship must be integrated into the developed device to ensure accurate measurements under various environments.

This paper first reviews international developments in nitrogen fluorescence-based remote α contamination detection technology and briefly describes our team's latest research. Through Monte Carlo simulations and experimental studies, we found that although α-induced nitrogen fluorescence is spatially concentrated, facilitating α source localization and analysis, its yield is extremely weak, emitting isotropically into space and being susceptible to environmental factors such as humidity and pressure. Achieving detection of radiation events producing only a few tens of photons per MeV at appropriate distances undoubtedly requires extremely low equipment noise and strict environmental control. Current research priorities for α contamination remote detectors remain reducing background light interference and improving fluorescence collection efficiency to enable effective α contamination detection under natural lighting conditions.

Practical implementation makes device development more complex. For engineering applications, achieving accurate detection of α-induced fluorescence in illuminated environments without background interference is crucial. For fluorescence detection, incorporating temporal information for coincidence measurements or appropriate data algorithms for automated processing is essential. For prototype development, beyond selecting appropriate photoelectric conversion devices, thorough investigation of field of view, pixel resolution, and detection distance is needed to determine optimal imaging performance. Future research must address several aspects, including but not limited to:

1. Investigating methods to improve fluorescence production efficiency, including inert gas purging;

2. Further optimizing filtering methods and electronic components to reduce noise impact on characteristic signals;
3. Investigating nuclide identification based on air fluorescence methods;
4. Developing imaging methods for coupled fields of multiple contamination sources in complex environments;
5. Establishing standards for remote α surface contamination measurement based on air fluorescence;
6. Investigating the feasibility of characterizing environmental samples using air fluorescence methods.

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Note: Figure translations are in progress. See original paper for figures.