Air Pollution Control at a DOE Facility

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Abstract
The Department of Energy (DOE) plutonium production program produced some of the greatest scientific and engineering accomplishments of all time. It is remarkable to consider the accomplishments of the Manhattan Project. The B Reactor on the Hanford Site, the first production reactor in the world, began operation only 13 months after the start of construction. The DOE nuclear production program was also instrumental in pioneering other fields such as health physics and radiation monitoring. The safety record of these installations is remarkable considering that virtually every significant accomplishment was on the technological threshold of the time.

One other area that the DOE facilities pioneered was the control of radioactive particles and gases emitted to the atmosphere. The high efficiency particulate air filter (HEPA) was a development that provided high collection efficiencies of particulates to protect workers and the public. The halogen and noble gases also were of particular concern. Radioactive iodine is captured by adsorption on activated carbon or synthetic zeolites.

Besides controlling radionuclide air pollution, DOE facilities are concerned with other criteria pollutants and hazardous air pollutant emissions. The Hanford Site encompasses all those air pollution challenges.

Introduction
The DOE facilities involved in the nuclear weapons business are now largely out of business. The West won the Cold War. Of course, there were costs to winning the War. For DOE and the U.S. citizens it is a legacy of sites contaminated with radioactive, hazardous, and mixed-wastes. Huge volumes of waste must be managed in a safe storage mode until acceptable waste treatment technologies are developed. Associated with managing and maintaining the waste tanks, the reprocessing canyons, the retired reactors, and other waste sites are air pollution issues.

The Hanford Site is one such DOE facility. The Hanford Site (Site) is located on 560 square miles in south-central Washington state. The area is characterized by bunch-grass, sagebrush vegetation in a semi-arid climate. About 6% of the Site has been disturbed or contaminated. Certain of the areas have played an important role in local Native American cultures. The Site is located on the banks of the last free-flowing stretch of the Columbia River in the United States. These issues contribute to the challenge of managing the Hanford Site, since there are diverse stakeholders interested in activities on the Site. These stakeholders are invited to help manage the Site. The public, the Indian Nations, and the state regulators have a very active role in deciding issues related to environmental compliance, clean-up, and future land use. This can lead to a policy of consensus management, which can lead to increased costs.

The effort to involve the stakeholders is not necessarily unwarranted, however. Recently declassified reports of human experiments with radioisotopes in the early days of nuclear research have been well documented in the news. Past releases of radionuclides from DOE sites have caused the "down-winders" to be concerned about health impacts. These events have only recently
been publicized. So a history of secrecy and past practices has created a feeling of distrust in the people and state authorities surrounding these sites. These attitudes contribute to the challenges of managing many DOE plutonium production sites.

Another communication problem faced by the DOE sites is the complex and technical nature of the activity. The radionuclide component of the equation poses some interesting issues, as the commercial nuclear industry knows. The industry is plagued by a science and terminology not understood by the general public. This makes risk communication very difficult. The situation is similar on the DOE sites. Terms like curies, roentgens, becquerels, and rem are difficult to describe in terms meaningful to the lay person. The nuclear industry's failure to adequately communicate risk is one reason for the nuclear industry's twilight in this country.

Two other changes are happening concurrently at the DOE Sites creating new air pollution control challenges. The Clean Air Act Amendments of 1990 are being implemented in the midst of drastic budget reductions. The funding issues for all environmental compliance activities have become very significant.

Air Emission Regulation
There are three components to the emissions from most of the DOE sites. There is the radioactive component from the fuel cycle facilities, the criteria pollutants from coal-fired steam generating units and fossil-fueled emergency generators, and the hazardous air pollutants from a variety of sources ranging from fleet vehicle maintenance and operation to paint shops and welding booths. At the Hanford Site, three separate agencies regulate the air emissions from the Site.

The Washington State Department of Ecology (Ecology) regulates the criteria pollutants under Washington Administrative Code (WAC) 173-400 and the hazardous air pollutants under WAC 173-460. The Washington State Department of Health, Radiation Protection Division (Health) regulates the radionuclide air emissions (WAC 246-247). In addition, the Environmental Protection Agency (EPA) Region 10 also regulates the radionuclide air emissions under 40 CFR Part 61, Subpart H. Washington State is the first state to receive partial delegation of 40 CFR 61 Subpart H delegation from the EPA. This delegation should help alleviate the confusion that can develop when more than one agency regulates one pollution pathway.

The State of Washington Department of Health Radiation Protection Division is very actively regulating airborne radionuclide emissions from the Hanford Site. The rules are based on the as low as reasonably achievable (ALARA) concept. The off-site dose limit is 10 mrem per year, however the ALARA rules give Health broad authority to regulate emissions. New construction and significant modifications of existing facilities must utilize best available radionuclide control technology (BARCT). All existing emission units and nonsignificant modifications must utilize ALARA control technology (ALARACT). BARCT and ALARACT determinations take into account other factors such as economic considerations of application of the control technology.

The Ecology and Health authority also overlaps to a certain degree in the area of airborne radionuclide emission regulation. Ecology has the statutory authority for setting the radionuclide standards but Health has the authority to enforce the standard. The Ecology/Health relationship has been worked out through a memorandum of understanding (MOU) between the two agencies, and generally works quite well with a few exceptions.

Particulate emission regulation is one area where the multiple regulatory agency role has caused some confusion. Ecology enforces a 20% general opacity emission limit. There have been attempts by Ecology to regulate the opacity emissions from radionuclide emission sources with multiple stages of HEPA controls. DOE maintains that controlling
opacity from these emission units is superfluous since the HEPA's adequately control any particulate that may contribute to opacity. Certainly, any visible particulate emissions from radionuclide emission sources will pose a public health risk apart from opacity.

The Hanford Site is also subject to a federal facility compliance agreement (FFCA). This FFCA governs the monitoring of radionuclide airborne emissions from point sources of air pollution. The national emission standard for hazardous air pollutants (NESHAP) governing non-radon radionuclide air emissions from DOE facilities (40 CFR 61 Subpart H) dictates that any emission point with the potential to cause an offsite dose to the maximally exposed individual of greater than 0.1 mrem/yr. must be continuously monitored according to specific EPA standards. These stacks are commonly referred to as major stacks. There are about 20 major stacks on the Hanford Site. Several of these stacks have been determined to be in compliance with the monitoring standard and others are currently being assessed to determine the NESHAP monitoring compliance status. The FFCA contains a strict schedule for stack assessments of the major stack monitoring systems and monitoring upgrades when the assessments identify deficiencies compared to the EPA standards.

The Hanford Site is subject to a prevention of significant deterioration (PSD) permit issued by the EPA in 1980. The PUREX chemical reprocessing plant and the uranium trioxide plant (UO3) were subject to NOx emission limitations and continuous monitoring when these plants were operating. The UO3 plant was shut down after a final cleanout run in 1993. The PUREX plant is in transition to shutdown. The PSD permit NOx limits are enforceable during the PUREX plant transition to shutdown activities, however, there is very little chance of measurable NOx emissions during this phase, since no fuel reprocessing is planned. The air operating permit program is also an air pollution control issue on the Hanford Site and at most DOE facilities. The Washington State air operating permit program was granted interim approval on December 9, 1995. All major sources then had 180 days to submit their air operating permit application. The Hanford Site Air Operating Permit Application (Application) was submitted to Ecology on May 26, 1995, with a complete application determination received on July 24, 1995. Under the MOU between Ecology and Health mentioned earlier, Ecology is the lead agency for air operating permitting purposes. The Hanford Site is arguably the facility in the state with the most complex interaction between the two regulatory authorities, making Application preparation even more challenging.

The Application was prepared in about one year, including a site-wide air emission inventory. The inventory team began field work in the spring of 1994. Actual application preparation did not begin until October, 1994. The Application contains descriptions of about 350 individual emission points. There are over a thousand additional insignificant emission units described categorically in the Application. This effort to categorize and provide minimal descriptions of insignificant emission units is consistent with the EPA guidance issued on July 10, 1995, two months after the Application was submitted. Frequent communications with the state permit writers allowed the Hanford Site to take advantage of numerous innovative solutions to describing the emissions from a very complex site.

Another major time and money saving initiative was the treatment of emission calculation documentation. The Hanford Site used example calculations for each major category of emission point, for example, combustion sources. Then all the emission points for that category were listed. This allows the permit writer to verify the calculation and emission factors. If they wish to verify individual emission point calculations, the permit writer is invited to review the emission inventory records. This methodology is also consistent with the EPA July 10 guidance.
One major area of concern for the Hanford Site is the outcome of the debate between Ecology and EPA over insignificant emission units. Ecology has several state-wide standards for all emission points, such as 20% opacity, which is part of the approved state implementation plan and therefore federally enforceable. EPA contends that these standards require some degree of monitoring, recordkeeping, and reporting for all emission units, even those categorically exempt from the air operating permit program. The outcome of this issue may have significant compliance cost consequences for the Hanford Site.

An additional component of radionuclide air pollution control involves the DOE orders. Before 1988 DOE was largely self-regulating (claiming sovereign immunity). A series of DOE orders were developed to control all aspects of facility operation. The DOE Orders in the 5000 series generally apply to environmental concerns. The DOE radionuclide dose limit to members of the public is 100 mrem/yr to the maximally exposed individual from all pathways. The orders often apply additional requirements on the facilities meant to assure compliance with the DOE dose limit. Some of the DOE orders, e.g. DOE Order 5400.5, which regulates the emission of radionuclides, are being codified in the federal register, presumably giving DOE civil and criminal enforcement authority over its contractors. DOE Order 5400.5 is being codified at 10 CFR 834.

**Air Pollution Control Technologies**

Radionuclide control has posed interesting control challenges. The level of control that was required has historically been some of the most stringent applied to industrial emissions, since these pollutants have historically been the contaminants of greatest concern.

As Figure 1 indicates the doses associated with Hanford radionuclide airborne emissions have been decreasing as plants and reactors have been shut down. The current doses are well below the 10 mrem/yr. annual NESHAPs limit. Certain control technologies are utilized to maintain the radionuclide emissions at current low levels. Since shutdown of the plutonium production facilities, airborne emissions of radioactive particulate matter is the primary off-site dose pathway. For airborne emissions of radionuclides BARCT has generally been determined to be high efficiency particulate air (HEPA) filters. The filter media is a disposable, extended media, dry-type
paper filter that has a removal efficiency of 99.97% for 0.3 \( \mu m \) particles. HEPA filters are composed of very fine (submicron) diameter fibers in a matrix of larger diameter (1 to 4 \( \mu m \)) fibers. Corrugated separators are interleaved with the pleats of the filter medium to space the pleats and add strength to the filter core. The filter core is sealed into a case. In-place filter tests are conducted with a polydispersed dioctyl phthalate aerosol in a size range of 0.1 to 3.0 \( \mu m \) and a light-scattering number mean diameter of 0.7 \( \mu m \). The procedure for conducting in-place testing of HEPA filters is described in the American Standard of Mechanical Engineers/American National Standards Institute N510 standard.

The treatment train for a nuclear facility may include other common air filters placed upstream of the HEPA filters. The pre-filters remove particles from the gas stream to protect the HEPA filters from dust plugging. If there is a potential for high humidity in the gas stream, there may be a demister to remove the moisture from the stream to prevent a wet stream from causing moisture plugging of the HEPA filters. Last in the treatment train are one or more banks of HEPA filters in series.

The noble gases and halogens, particularly \(^{131}\)I are no longer an issue on the Hanford Site. Since the N-Reactor was shut down in 1989 and no further fuel reprocessing is occurring, there is no longer any generation of these contaminants.

The noble gases such as xenon and argon have physically decayed away. In the past they were treated by hold-up to allow some decay before discharge to the atmosphere. The radioactive iodines were removed using activated carbon adsorber cells. For example, the Hanford production reactor filter system consisted of 4 compartments. Each compartment consisted of 36 moisture separators, 36 HEPA filters in a 4 x 9 array, and 36 pleated-bed charcoal adsorber cells.

Another area important from an air pollution standpoint are the emissions from the coal and oil-fired steam generators operated on the Site. These units provide process and space-heating steam for the chemical separations plants and the fuel fabrication and testing areas.

Figure 2 shows the criteria pollutant emission rates from the steam generating plants. The 200 Area coal-fired steam generating plants were constructed during the 1940's. These boilers are currently grandfathered from the new source performance standards (NSPS) for steam generating plants and PSD regulations. No major modifications since the new source performance standards have occurred that would have required new control technologies. Baghouses were installed on the two coal-fired plants in the mid-1970's to comply with the general opacity limit. These two plants alone account for about 63% of the \( \text{SO}_2 \) emissions, 85% of the NOx, 16% of the particulate matter, and 96% of the CO emissions from the Hanford Site. The relatively low contribution of particulate matter to the site emissions compared to the other pollutants demonstrates the effectiveness of the baghouses. Given the ages of these plants, any new regulations that may require retrofit best available control technology (BACT) or continuous emissions monitoring will, in all likelihood, cause their shutdown. In fact, studies are underway now to determine replacement steam generation requirements and possible solutions.

The other air pollution regulation that affects the Hanford Site is the State of Washington Department of Ecology toxic air pollutant (TAP) regulations (WAC 173-460). These regulations apply to all new or modified sources of TAPs. Several of the new treatment and waste handling facilities at Hanford have TAP permits. The most significant pollutants from the facilities that have been permitted are volatile organic compounds.

The TAP regulations have another variation of BACT known as T-BACT, i.e. toxic - best available control technology. New toxic air pollutant emission sources are assessed to
determine if the construction or modification will result in an increase of toxic air emissions. The list of toxic air pollutants is defined in the regulations. There are about 700 toxic air pollutants listed in WAC 173-460.

The regulations provide small quantity emission rate (SQER) tables and acceptable source impact levels (ASIL). The ASIL's are receptor concentrations determined through dispersion modeling. New construction or modifications are assessed against the ASIL's and SQER emission limits to determine the need for either of two levels of control; either T-BACT or toxic - reasonably available control technology (T-RACT). If a modification results in an increase of TAP's and is not otherwise exempt and falls within the purview of the regulations, T-BACT must be applied to control those pollutants that represent an increase in emissions. For TAP emissions that stay the same or decrease, T-RACT must be applied for those pollutants. For certain source categories T-BACT is defined in the regulations, e.g. perchloroethylene and petroleum solvent dry cleaners and solvent metal cleaners.

An example of the control technology requirements is the T-BACT for solvent metal cleaners. The control technology requirements include the following:

- A cover for the solvent tank that must remain closed except when processing work in the degreaser.
- A drain system for cleaned parts that returns drained solvent to the tank.
- For cold solvent cleaners, a freeboard ratio of greater than or equal to 0.75.

Vapor degreasers shall have:

- A high vapor cutoff thermostat with manual reset.
- For degreasers with spray devices, a vapor-up thermostat which will allow spray operations only after the vapor zone has risen to the desired level.
- Either a freeboard ratio greater than or equal to 1.00 or a refrigerated freeboard chiller.

Conveyorized degreasers also must be equipped with certain equipment.

Solvent degreasers must also meet various operational requirements to reduce solvent leaking and evaporation.
The Washington State TAPs regulations fulfill many of the requirements to comply with the construction, reconstruction, and modification requirements of Section 112(g) of the 1990 Clean Air Act Amendments. The EPA, in its November 9, 1995 Federal Register notice granting interim approval for the Washington air operating permit program, allowed that the TAPs regulations covers most situations where section 112(g) would apply. The EPA intends to publish a separate Federal Register notice to propose approval of Washington's TAP program. Final approval will represent EPA delegation of section 112(g) authority to the State.

Conclusion
Air pollution control at a DOE facility can be a very complex issue. The variety of the facilities and operations and multiple regulatory authorities that span the range of activities from vehicle maintenance to mixed-waste management provide challenges not encountered at many other industrial facilities in the country. When current budgetary constraints are factored into the equation, the air pollution compliance issues are even more challenging.

References:


Radiation Protection of the Public and the Environment. DOE Order 5400.5. United States Department of Energy


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