2. GASEOUS WASTE (NPWT 1978)

During operation, nuclear power reactors generate radioactive fission products, among them xenon and krypton gases. A portion of these will be released to the coolant when there are fuel cladding defects. Because the gases are not completely soluble within the coolant, they are available for release to the environs. To limit the radiation exposure to the public, off-gas treatment systems for removing these volatile fission products are installed at nuclear power plants. Activity releases will occur from several points. However, the following discussion is limited to the treatment of the steam jet air ejector (SJAE) of a boiling water reactor (BWR) and the gas stripper of a pressurized water reactor (PWR), since these are the major sources of gaseous release from these plants. But, after the installation of treatment equipment on these streams, they represent only a small fraction of the total plant radioactive release. In setting the activity release design criteria for the entire off-gas system, the relative contribution of the SJAE or gas stripper treatment system should be considered with respect to the total plant release. A study of offsite doses will reveal that once the off-gas system has been installed, greater dose reduction can be achieved by reducing the activity releases from sources other than the SJAE or gas stripper treatment systems. Thus, the design activity release from these off-gas treatment systems should not be determined independently.

Gaseous radioactive source terms (NPWT 1978)

The methodology used in the calculation of liquid and gaseous radioactive source terms for LWR nuclear power plants is provided. The parameters given are consistent with those used in the GALE Code. The model discussed for PWRs is intended for use with those using zircalloy clad fuel, continuous purification of the primary coolant, and recirculating U-tube or once-through steam generators. The model discussed for BWRs is intended for use with those using zircalloy clad fuel and continuous demineralization of the reactor coolant. These models, instructions for their use, and the bases for all parameters and release rates specified in this chapter are available in Regulatory Guides published by the NRC. The parameters used herein were derived from reactor operating experience where data were available. Releases of radioactive materials in gaseous effluents include both releases from process systems and releases through ventilation pathways. Process system effluents contain radioactive materials as a result of stripping or venting radioactive gases from process streams. Ventilation pathways contain radioactive materials as a result of radioactive process fluid leakage into building air spaces serviced by the ventilation systems.

BWR gaseous source term model

The principal BWR gaseous release points (F-1) are the main condenser evacuation system, turbine gland seal system, mechanical vacuum pump exhaust and ventilation system exhaust from containment, auxiliary, turbine and radwaste buildings. Other potential sources have been evaluated and found to have a negligible effect on the overall source term.

The Main Condenser Evacuation System is the principal release point in a BWR before treatment. Noble gases and radioiodine, carried over with the main steam flow from the reactor vessel, are normally vented from the main condenser through an air ejector. If not treated, the effluent releases would amount to approximately 1.5E+6 Ci of noble gases (assuming 30 minutes, decay) and 5 Ci of iodine-131 per 3400-MW(t) reactor. The radioactive gases leaving the air ejector are dispersed in a gaseous stream with the following approximate proportions for a 3400-MW(t) reactor at standard temperature and pressure: flow rate (l/min): hydrogen 3400, oxygen 1700, air 850, water vapour 710. The hydrogen and oxygen are present in stoichiometric proportions from radiolytic decomposition of water. The amount of air is based on a three-shell main condenser and 10 cfm (280 l/min) air inleakage per main condenser shell. To reduce the volume of gas, the oxygen and hydrogen are recombined and the water vapour is condensed out of the gas stream leaving approximately 30 cfm (850 l/m) of noncondensables to be processed. Effluent gases from the main condenser air ejector are normally delayed prior to release to permit the decay of short lived radionuclides. The most common process proposed for treating off gases from the main condenser air ejector is dynamic adsorption on charcoal.

Steam is supplied to the turbine gland seal system to prevent air inleakage to the turbine through the turbine shaft seal. Air inleakage to the gland seal annulus and noncondensable gases present in the steam are vented through the gland seal condenser vent to the environment. If the steam supplied to the turbine gland seal system is withdrawn from the main steam system, radioactive gases present in the main steam will be released through the gland seal condenser vent. When main steam is used, the gland seal vent gases are normally passed through a holdup line to provide decay of the short-lived radionuclides. The flow rate through the turbine gland seal system is approximately 1.5E+4 lbs/h (6800 kg/h) of sealing steam (0.1 % of the main steam flow rate) and 600 ft^3/min (1.7E+4 l/min) of noncondensable gases (radiolytic oxygen and hydrogen and air inleakage). Steam condensation in the gland seal condenser results in the partitioning of iodine between the liquid phase and noncondensable gaseous phase. For source term calculations, a partition factor of 0.01 is assumed, which means 1 % of the iodine remains in the gaseous phase and 99% enters the liquid phase. Additional iodine removal may be provided by processing the effluent gases through charcoal adsorbers prior to release. Prior to treatment, the annual releases from the gland seal condenser vent considering two minutes holdup prior to release are approximately 6800 Ci/y for noble gases and 0.049 Ci/y for iodine-131. It is common in present BWR designs to produce the turbine gland seal steam in an auxiliary boiler using feedwater condensate. Such systems are said to use "clean steam". "Clean steam" is a term applied to steam supplied from a source with sufficiently low activity that the effect on the gaseous source term is negligible (resulting in noble gas releases of less than 1 Ci/y and I-131 releases of less than 1E-4 Ci/y). The radioactive source term is negligible for systems supplying clean steam to the turbine gland seal system.

Mechanical vacuum pumps are used to establish main condenser vacuum during startup and to maintain a slightly negative pressure in the main condenser during shutdowns. The mechanical vacuum pump effluent contains radioactive materials due to residual activity remaining in the main condenser following reactor shutdowns. The principal constituents in the mechanical vacuum pump effluent are Xe-133, Xe-135 and I-131. Since the mechanical vacuum pump flow rate may be high, treatment processes are normally not proposed for reducing noble gas releases from the mechanical vacuum pump. However, charcoal adsorbers may be installed to reduce radioiodine releases from the mechanical vacuum pump. If no treatment is provided, the annual releases from the mechanical vacuum pump are approximately 2650 Ci/y of noble gases and 0.03 Ci/y of iodine-131.

BWR ventilation system releases

Buildings housing components used to process radioactive material can be expected to release radioactive materials in their ventilation air effluents due to leakage from components into building air spaces. The radioactive materials released will include noble gases, radioiodine, tritium, and radioactive particulates. The buildings expected to be of significance to the source term calculations (in order of significance) are the turbine, containment, auxiliary, and radwaste buildings. Tritium releases from the fuel building may be significant; however, tritium releases are considered on a "per plant" rather than "per building" basis in the gaseous source term calculations. Radioactive releases from other buildings are considered to be negligible.

The gaseous tritium source term is based on the reactor power level and on the quantity of liquid discharged from the plant. The total quantity of tritium available for release through all effluent pathways is considered to be 0.025 Ci/MWty. Thus for a 3400-MW(t) plant, the estimated total tritium release would be approximately 85 Ci/y. The tritium release is calculated by multiplying the volume of liquid released by the nominal tritium concentration in the BWR coolant streams (0.01 mCi/l). The difference between the total tritium release and that released through liquid pathways is assumed to be released in gaseous effluent.

Table 13 Comparative releases from the BWR main condenser air ejector (Ci/y/3400 MWt reactor).

RI	Kr-85m	Kr-85	Kr-87	Kr-88	Xe-133	Xe-135m	Xe-138	I-131	I-133
T	4.5h	10y	76m	2.8h	5.3d	15m	14 m	8 d	21 h
FY	1.31	1.31	2.56	3.57	6.7	6.5	6.76	2.89	6.7
BC	8E+4	290	2E+5	3E+5	1E+5	1E+4	4E+5	5	2.1
C	80	290	<1	5	460	<1	<1	<a	<a
CD	22	280	72	76	13	3	90	3E-3	0.01

a = 1E-4, FY = Fission Yield - %, BC = Base case (30 min holdup), C = Charcoal (24 ton system operating at -18 C, -29 C dew point, 850 l/min air leakage, 42 d holdup for Xe and 1.8 d holdup for Kr), CD = Cryogenic Distillation (based on distillation partition factor of 0.0001 for Xe, I and 0.00025 for Kr and holdup time of 90 d for gases collected).

PWR gaseous release points

The principal PWR gaseous release points are the waste gas processing system exhaust, steam generator blowdown flash tank vent and exhausts from the main condenser evacuation system, containment turbine and auxiliary buildings. The releases from the containment building, auxiliary building, and waste gas processing system are calculated based on the concentration of radioactivity in the primary system. Releases from the steam generator blowdown flash tank vent, the turbine building, and the main condenser evacuation system exhaust are calculated based on the concentration of radioactivity in the secondary system, which is a function of the activity in the primary system.

The rate of gaseous release to the primary coolant is a function of the plant power level and the integrity of the fuel cladding. The PWR-GALE Code calculations assume that 0.12% of the equilibrium fission product inventory in the fuel is released to the primary coolant. The rate of noble gas and iodine release to the primary coolant is governed by the rate of gaseous diffusion through the fuel matrix and cladding. Empirical constants termed escape rate coefficients" are used in conjunction with the "% cladding defects" to calculate the fission product release rate to the primary coolant. The following escape rate coefficients are typical of those used for PWR source term calculations (sec-1) : Xe, Kr 6.5E-8, I 1.3E-8. The steady-state gaseous activity in the primary coolant is primarily a function of the release rate discussed above and of two principal removal mechanisms; radioactive decay in the primary coolant and removal by gaseous stripping. Low gaseous stripping rates (stripping the shim bleed at approximately 1 gpm, 3.8 l/min) result in long radionuclide residence times in the primary system. Raising the gaseous stripping rate (stripping the total letdown flow at approximately 75 gpm, 290 l/min) decreases the radionuclide residence time in the primary system, thereby reducing the amount of radioactive decay occurring while the radionuclides are in the primary system. Comparison of the effects of stripping rates shows that high stripping rates result in lower gaseous activities in the primary coolant, thereby lowering the noble gas source term for release pathways which are based on primary coolant leakage (e.g., containment and auxiliary building vents). The waste gas processing system input rate is increased by high stripping rates; however, it can compensate for the increased gaseous input rate by increasing the capacity of the gaseous holdup system. If charcoal delay is used in the waste gas processing system, the holdup time is calculated using the equation and dynamic adsorption coefficients. For source term calculations, the gaseous flow 0.5 ft^3/min (14 l/min).

PWR ventilation system releases

Leakage from the primary system will enter the containment and auxiliary building ventilation effluent air streams. Releases from the reactor containment building are calculated assuming four purges per year and a 1000 cfm (3800 l/min) continuous purge during operation. Radioactivity present in the containment atmosphere is due to primary coolant leakage to the containment building from primary system components. It is assumed that 1% of the noble gas inventory and 0.001% of the iodine inventory in the primary coolant is released to the containment atmosphere daily as a result of system leakage and component venting. These parameters were derived from measurements of containment building airborne concentrations and primary coolant activities at operating PWRS. Gaseous releases from the containment building may be altered by several variables. The designer may elect to use periodic purges in place of the continuous purge or to use a lower continuous purge rate which will allow greater radioactive decay to occur in the containment building prior to release. Internal recirculation systems which recirculate the containment ventilation air through HEPA filters and charcoal adsorbers prior to release may be used to lower the activity levels in the containment building prior to purging. If internal recirculation systems are used, assume 16 hours recirculation, 70% mixing efficiency, and DFs of 10 and 100 for charcoal adsorbers and HEPA filters, respectively. In addition, the containment building ventilation air may be processed through charcoal adsorbers and HEPA filters for radioiodine and radioactive particulate removal during release. The use of any or all of the above methods may be factored into the source term calculations.

Auxiliary building gaseous source term calculations are based on 160 lbs/day (73 kg/day) of primary coolant leakage to the auxiliary building. It is assumed that all of the noble gases and 0.75% of the radioiodine present in the primary coolant are released to the auxiliary building ventilation air. Radioiodine and radioactive particulate releases may be reduced by means of charcoal adsorbers and HEPA filters. The available treatment processes for ventilation releases are not practicable for noble gas removal.

PWR secondary system sources

Releases from the main condenser air ejector, steam generator blowdown system vent, and the turbine building ventilation system are a function of the secondary coolant activity. The secondary coolant activity is based on a primary system to secondary system leakage rate of 100 lb/day (43 kg/day) and one of several cleanup mechanisms. First, for systems using phosphate chemistry, the steam generator blowdown rate is assumed to be 0.06% of the main steam flow rate. For phosphate chemistry systems the steam generator blowdown is the major mechanism for removing activity from the secondary system. Second, for systems using volatile chemistry it is assumed that the steam generator blowdown rate is 0.5% of the main steam flow rate. Since approximately 35 % of the feedwater is "forward pumped" as extraction steam and moisture removed in the moisture separators and, therefore, bypasses the condensate demineralizers, it is assumed that 65% of the steam generator feedwater is passed through a condensate demineralizer. For volatile chemistry systems both the steam generator blowdown and the condensate demineralizer systems contribute to the cleanup of the secondary coolant. Third, for once-through steam generator systems, the secondary coolant activity is calculated based on 65% of the steam generator feedwater being processed through a condensate demineralizer blowdown is not used on once-through steam generators.

Turbine building ventilation system source terms are based on 1700 lbs/h (770 kg/h) of steam leakage to the turbine building. The noble gas source term is independent of the type of steam generator used since essentially all of the noble gases are carried over with the main steam. The iodine source term is dependent upon the type of steam generator used. Approximately 5% of the primary coolant iodine is in a volatile form which is completely volatilized upon entering the steam generator. Recirculating U-tube steam generators have a liquid reservoir where nonvolatile iodine species tend to concentrate. Then they are carried over with the main steam as a result of moisture carryover. The assumption that 1% of the iodine in the steam generator is carried over with the steam is reasonable for recirculating U-tube steam generator source term calculations. Source term calculations for once-through steam generators should be based on 100% iodine carryover.

Radioiodine entering the steam phase in the steam generator is, for the most part, carried over to the main condenser. The iodine entering the main condenser will consist of both volatile and nonvolatile species. Nonvolatile species will enter the liquid phase with the condensing steam and will not contribute to the gaseous source term. Volatile species will partition between the condensing steam and the noncondensable gases in the main condenser. Approximately 15% of the volatile species entering the main condenser with the steam will be released through the air ejector exhaust. The remaining 85% will enter the liquid phase and return to the steam generators with the feedwater.

It is assumed that the volatile radioiodine species that are not removed through the main condenser air ejector are reduced to a nonvolatile form before they return to the steam generator due to the secondary system chemistry. For this reason, most of the iodine in the liquid phase in the steam generator is in nonvolatile form. Consequently, the radioiodine present in the steam generator blowdown liquid is also nonvolatile. Similarly, the noble gas concentration in the steam generator blowdown liquid is very low since most of the noble gas is carried over with the main steam flow in the steam generator. As a result, noble gas releases from the blowdown flash tank vent are negligible and iodine releases will only occur as a result of moisture entrainment in the flashing steam. Approximately one-third of the blowdown stream flashes to steam in the flash tank. Based on 85% steam quality, approximately 5% of the iodine in the blowdown stream may become airborne. If the blowdown flash tank is vented through a condenser or if the blowdown liquid is cooled below 212 F (100 C) to prevent flashing, iodine releases from the blowdown system will be negligible.

If charcoal adsorbers or HEPA filters are used to treat any of the above sources, the effluent source terms are reduced by factors of 10 and 100 for radioiodine and radioactive particulates, respectively. The particulate concentration in the secondary coolant is sufficiently low that particulate removal is generally not required for secondary system sources. All of the iodine and noble gases present in the turbine building steam leakage are assumed to be released with the ventilation air. PWR turbine buildings are relatively open structures which are not amenable to charcoal adsorption or HEPA filtration.

The gaseous tritium source term is based on the reactor power level and on the quantity of liquid waste released, excluding secondary system wastes. The total quantity of tritium available for release through all effluent pathways is considered to be 0.4 Ci/MW(t)y. Thus, for a 3400-MW(t) PWR plant, the estimated total tritium release would be 1 360 Ci/y. The annual tritium release is estimated by multiplying the calculated volume of liquid released from primary system sources by the nominal tritium concentration in PWR primary coolant (1.0 mCi/l). The difference between the total tritium release and that released through liquid pathways is released in gaseous effluents.

Table 14 Comparative releases from the PWR waste gas processing system (Ci/y/3400 MWt reactor).

RI	Kr-85m	Kr-85	Kr-87	Kr-88	Xe-133	Xe-135	Xe-138	I-131	I-133
T	4.5h	10y	76m	2.8h	5.3d	15m	14 m	8 d	21 h
NT	1E+4	300	8E+3	2E+4	2E+5	3E+4	4900	3	4.4
C	<1	300	<1	<1	18	<1	<1	<a	<a
PS	300	300	72	<1	89	<1	<1	<a	<a

NT = No Treatment, C = Charcoal (72 d holdup for Xe and 4 d holdup for Kr), PS = Pressurized Storage 60 d.

Other source terms

In addition to the radioactive material releases discussed above, small quantities of C-14 and Ar-41 will be released in gaseous effluents. Carbon-14 is produced in a thermal neutron reaction with oxygen-17 [O-17(n,alpha)C-14] in the water flowing through the core. Approximately 9.5 Ci/y of C-14 is expected to be produced in a 3400 MW(t) BWR. The C-14 will be released through the main condenser air ejector. The C-14 source term for a 3400-MW(t) PWR is expected to be approximately 8 Ci/y released primarily through the waste gas processing system. Argon-41 is formed by neutron activation of stable naturally-occurring argon-40 in the air surrounding the reactor vessel. The argon-41 source term for both BWRs and PWRs is approximately 25 Ci/y with the BWR releases occurring during purging of the reactor drywell and the PWR releases occurring from purging the containment building.

Table 15 Comparison of number of gaseous atoms created (NC) in a reactor with those released (NR) when no off-gas treatment used (per GWy).

RI	Kr-85	Kr-88	Xe-133	Xe-138
NC	8E+19	1E+20	2E+20	2E+20
NR : BWR	1E+9	4E+9	7E+10	5E+8
NR : PWR	2E+8	4E+8	1E+11	6E+6

Note : these calculations were made assuming total efficiency of 33% and that 1 GWy = 3E+21 fissions.

Gas treatment technologies

Radiolysis of the cooling water in the reactor vessel, generates gaseous hydrogen and oxygen. These gases, along with the fission product noble gases released from the fuel are carried by steam to the condenser. These gases plus air inleakage are removed from the main condenser by the SJAE. Several process systems, ranging from simple storage to cryogenic distillation, have been developed to remove fission product gases from the SJAE exhaust (F-15).

Compressed gas storage

Compressed gas storage systems achieve a reduction in activity releases by simply storing the fission product gases under pressure in large tanks and allowing them to decay (F-5). The volumetric gas flow rate from the SJAE is reduced by using a recombiner subsystem which removes the excess hydrogen and oxygen. Removal of these gases reduces the SJAE effluent flow rate to about 20 % of the influent flow rate. The gas is then passed through a 10 minute holdup pipe where decay of the short half-life fission products such as Kr-89 and Xe-137 takes place. A HEPA filter located downstream of the holdup pipe removes the particulate daughter products of these noble gas isotopes (especially the long-lived Sr-89 and Cs-137). Since the gas at this point consists mostly of air saturated with water vapour, drainage is provided for both the holdup pipe and the HEPA filter. If building layout permits, the HEPA filter vessel can drain to the holdup pipe. After passing through the HEPA filter, the gas is compressed to 14-21 kg/cm^2, gage (200 to 300 psi), and directed to one of several storage tanks. Because of the high operating pressure, it is not feasible to design the system for hydrogen explosion containment either at the compressor or beyond it. The probability of a hydrogen reaction must therefore be reduced by proper design. To provide greater assurance that hydrogen gas does not enter the downstream portion of the system, redundant hydrogen gas analysers are installed after the off-gas condenser. The compressed gas system is capable of achieving the desired off-site dose reduction. However, the tank volumes required, while much less than that of a delay pipe, are still excessive. Although systems of this type are now in operation, compressed gas systems are not being proposed for new plants.

Gaseous holdup in the waste gas processing system is normally accomplished by storage in pressurized storage tanks or by selective adsorption on charcoal (charcoal delay system).

There are two general types of pressurized storage systems, i.e., high pressure storage tanks and nitrogen recycle systems.

High pressure storage tank systems (consider four tanks) generally operate in the following manner:

- one tank receives waste gas from the gas stripper. For source term calculations with base-loaded operation, it is assumed that the annual average gaseous flow rate to the gas tank is 140 ft^3/d (4E+3 l/d). The gaseous collection time is calculated as follows:

$$Tf = PV/F \qquad (1)$$

where Tf = time to fill tank (days), P = 70% of design storage pressure (atm.), V = tank volume (ft^3) and F = annual average gaseous flow rate to gaseous waste system.

- one tank is held in reserve for contingencies (e.g. back to back shutdowns).

$$Th = PV(n-2)/F \quad (2)$$

where Th = holdup time (days), P, V = same as above, and n = number of tanks in system. The factor (n-2) adjusts the total storage volume (nV) for one tank filling and one tank assumed held in reserve. The PWR-GALE Code calculates the effective radionuclide decay occurring during filling based on Tf. If the total decay time [effective decay time during filling plus the holdup time (Th)] exceeds 90 days, the releases are calculated based on 90 days. This is analogous to the BWR discussion on cryogenic distillation. After 90 days, only K-85 remains as a significant isotope. The above equations assume that all of the gases stripped from the coolant are stored for decay. If a recombiner is used to remove the hydrogen from the waste gas stream, the volumetric gaseous flow rate is very small and the above equations will underestimate the holdup time. For systems using recombiners, holdup time should be adjusted on a case by case basis based on the calculated input flow and proposed mode of operation.

Charcoal delay

The charcoal delay system utilizes the dynamic adsorption of Kr and Xe to remove these radioactive gases from the process off-gas prior to release. Experimental data indicated that charcoal could be used since it exhibits a dynamic adsorption of both these noble gases from air. The required holdup time (i.e., removal efficiency) of a charcoal bed is a function of the gas flow rate, mass of charcoal, and dynamic adsorption coefficient, K. The value of K, in turn, depends on the concentration of fission product gases, system operating pressure, system operating temperature (F), and moisture content of the charcoal. An equation for the charcoal holdup time is :

$$T = 2.5E\text{-}5\ MK/F \quad (3)$$

where T = holdup time (days), K = dynamic adsorption coefficient (cm^3/g) M = mass of charcoal (kg), and F = flowrate through system (cfm).

The following values are recommended for the dynamic adsorption coefficients:

Table 16 Dynamic adsorption coefficients.

Operating Temperature	- F	77	77	0
Dew point	- F	45	0	-20
Kr	cm^3/g	19	25	105
Xe	cm^3/g	330	440	2410

Dynamic adsorption coefficients based on laboratory tests and operating performance vary from 30 to 10000 cm^3/g and they are strongly dependent on temperature.

Ambient charcoal

Selective adsorption of fission products by charcoal can be used to reduce radioactive releases from the off-gas system. In a PWR, gases leaving the surge tank (F-16) are compressed to 14 kg/cm^2 and are passed through an after-cooler to remove heat and humidity. Then the pressure is reduced to 1 kg/cm^2 and the gas is passed trough the charcoal bed. For the flow rates associated with a PWR, 1.4 tons of charcoal are required for 30 days of Xe holdup. Prior to discharge, the gases are filtered to prevent charcoal fines from being released. In a BWR (F-6), after leaving the recombiner portion of the system, the gas can either enter a 10-minute delay line to allow for decay of the short half-life fission products, or may, as an alternative, pass through a small charcoal bed which will hold the fission product gases long enough to remove those having short half lives. To increase the adsorption efficiency of the charcoal, any water vapour remaining with the gas is extracted by a moisture removal subsystem. The charcoal is contained in several tanks operated in series downstream of the moisture removal equipment.

Refrigerated charcoal

Decreasing operating temperature increases the adsorption coefficient thus decreasing the volume of charcoal required to achieve a given holdup time. Reduction of the bed temperature from ambient to -18 C (0 F), increases the adsorption coefficient by a factor of three, and thus reduces the required charcoal volume by the same factor. Cost savings realized from smaller charcoal volumes must be balanced against the added cost of refrigeration equipment and insulation for the charcoal unit. Prior to entering the charcoal guard bed (F-9), the off-gas stream is processed through a desiccant dryer to remove water vapour which would otherwise freeze out in the nominal -18 C (0 F) vault. Removal of water vapour from the off-gas stream also increases the ability of the charcoal to remove fission product gases. The vault temperature of -18 C (0 F) is maintained by a mechanical refrigeration unit which also cools the influent gas stream. The cooled gas then enters the charcoal beds where Kr and Xe are removed prior to discharge. Off-site dose reduction factors for a refrigerated charcoal system can be increased as necessary by adding more charcoal. The system has the advantage of large dose reduction factors, comparatively low capital costs, and relatively small space requirements. Charcoal adsorption systems have different holdup times for Kr and Xe since the dynamic adsorption coefficient for Xe is 18 times higher than that for Kr. For a fixed quantity of charcoal, the Xe holdup time is therefore 18 times longer than for Kr. Fortunately, except for Kr-85, the Kr isotopes have shorter half lives than the Xe isotopes. A charcoal delay system which provides a satisfactory Xe dose reduction will also supply a similar dose reduction for Kr.

Cryogenic charcoal

The cryogenic charcoal uses the principle of lowering the temperature to reduced the charcoal volume required. It is capable of removing essentially all of the Kr and Xe from the influent gas. The saturated bed is regenerated by passing heated nitrogen gas through the bed and causing Kr and Xe to desorb. The gases are then compressed and stored in gas cylinders prior to release. Cycles times for cryogenic charcoal are on the order of 240 h. Essentially all the fission product gases can be removed from the plant off-gas stream by a cryogenic charcoal system. After the fission product gases are removed from the primary coolant, they are temporarily stored in a surge tank which allows batch operation of the system. First (F-18), trace quantities of oxygen and ozone are removed in the recombiner by reaction with excess hydrogen. In this way, ozone is prevented from building up in the cryogenic portion of the system. Next the gas is cooled and passed through a moisture separator and desiccant dryer which serve to keep ice crystals from plugging the cryogenic charcoal bed as it adsorbs the fission product gases. This charcoal bed is maintained at a temperature of approximately -170 C (-275 F) by using the nitrogen gas that boils off a liquid nitrogen bath to remove the heat of adsorption. The -185 C (-300 F) liquid nitrogen bath is used to cool the decontaminated effluent gas from the cryogenic charcoal bed prior to passage through the regenerative heat exchanger which returns it to ambient temperature. This process stream, which is essentially free of noble gases, is regulated at a gage pressure of 0.7 kg/cm^2 (10 psig), monitored for radioactivity and passed through a filter to remove particulates before venting to the atmosphere. When the charcoal adsorber bed reaches saturation as indicated by radiation monitors which detect the break-through of Kr, the bed is isolated from the system and regeneration is started. By use of a controlled heat source, the noble gases and any adsorbed carrier gas are desorbed and then stored temporarily in an accumulator tank. The tank provides surge capacity in the event that the gas desorption rate exceeds the capacity of the diaphragm compressor to transfer the gases to storage bottles. When the desorption is complete, as indicated by a bed temperature of 150 C (300 F), the system is purged with nitrogen gas. At this time, the desiccant dryer is also regenerated by heating and purging with clean nitrogen. The nitrogen purge gas is stored in the bottles with the noble gases. A cryogenic charcoal system for a PWR plant is extremely compact. The entire system, except for the surge tank, control panel, and liquid nitrogen storage tank, can be contained on a 2.7-m x 2-m (9-ft x 7-ft) skid which is 3-m (10-ft) high.

Cryogenic distillation

Because they are chemically inert, the noble gases are best removed from the off-gas system by physical means. Cryogenic distillation is still another such method. The Kr and Xe are condensed out of the gas stream as it passes through a distillation column operated at very low temperatures (-152 C for Kr and -108 C for Xe). The degree of noble gas removal is a function of distillation column size. Off-gas from the recombination system is first compressed (F-11) and then passed through a prepurifier which removes carbon dioxide and water vapour by freezing them out on the heat exchanger surface. The heat exchanger coolant is nitrogen gas vented from the cryogenic distillation volume. Effluent from the prepurifier is introduced into the cryogenic distillation column. As the process temperature is lowered from ambient conditions, the gases begin to liquify, with the amount of liquefaction of each gas depending upon its boiling point and vapour pressure.

Table 17 Normal boiling points (BP) for the gases which comprise the off-gas stream are.

Gas :	H_2	Ne	N2	Ar	O	CH4	Kr	Xe	CO	H_2O
BP-C:	-253	-246	-210	-186	-183	-164	-152	-108	-79	100

Oxygen and nitrogen, which account for 99 % of the gas flow after removal of the CO and H_2O by the freeze-out heat exchangers, have boiling points well below those of Kr and Xe. The condensed Kr and Xe are collected in a sump located in the distillation column. When the sump becomes full, the liquified Kr and Xe are transferred to a gas cylinder for storage and decay prior to eventual release of the Kr-85. Unless these gases are stored indefinitely, all the Kr-85 released from the fuel will be discharged to the environment. A sump purification loop limits concentrations of ozone and hydrocarbons to below explosive levels. The ozone is formed from the oxygen present in the high radiation field. Ozone concentration can also be limited by eliminating oxygen from the influent stream instead of hydrogen. The oxygen is removed by adding hydrogen to the off-gas flow upstream of the recombiner. The effluent concentration of oxygen is reduced to trace levels (on the order of a few ppm) by the addition of a secondary preheater, recombiner, and condenser which are sized for the air inleakage flow rate. Small activity releases can occur from several points in a cryogenic distillation system. The small fraction of the inlet activity which is not condensed in the distillation column will be vented from the top of the column. This release represents only a small %age of the total release from the system. The major sources of radioactive release are valve stem leakage and the venting of regeneration gas. These releases are included in the overall system decontamination factors of 4000 for Kr and 10000 for Xe. Despite the existence of several release points, the total activity releases are very small as indicated by these large decontamination factors. Distillation systems are capable of very low release rates, but when compared to a refrigerated charcoal system, the gained incremental reduction in release is an insignificant part of the total plant release.

Aerosol filtration (IAEA 1980)

The HEPA (high efficiency particulate air) filter (F-5.13) had its origin as a military countermeasure during WW 2 and was greatly improved during the next decade by the U.S. Naval Research Laboratory's development of high efficiency all-glass fiber filter paper that substantially exceeded then current filter performance standards. This all-glass fiber filter paper was pleated between corrugated spacers that held the folds of the paper about 9 mm apart on the up and downstream sides and the pack was sealed into an open-ended wood or metal box to form the filter cartridge. Using this paper and cartridge design, manufacture of noncombustible, all-mineral HEPA filters for service in hazardous locations became a reality. U.S. and U.K. filters were very similar and they became the mainstay of the nuclear industry for the past three decades, experiencing only minor modifications in materials and construction. This is understandable as these filters reached a state of near perfection with respect to retention of submicrometer particles when filter manufacturers found ways of improving their assembly techniques to the degree that they were routinely able to turn out filters that exceeded required particle retention efficiency by an order of magnitude, i.e. from 99.97% efficiency to 99.997%. In addition, the filters exhibit notable resistance to chemicals, flame, high temperature, and radiation.

To an important degree, the establishment of USAEC Quality Assurance (QA) Filter Test Stations in 1960 made it imperative for filter manufacturers to institute their own rigid quality control practices to avoid product rejections. By 1978, the rejection rate had declined to a point where the U.S. Nuclear Regulatory Commission was willing to forego QA Filter Test Station review for filters intended for use as engineered safety feature (ESF) systems in commercial nuclear power plants on the basis that the marginal increase in the reliability of tested filters no longer justified the addition of 30% to filter costs. There is currently much interest in prolonging the service life of HEPA filters with the use of low resistance prefilters, some of which have attained greatly improved particle retention characteristics by the application of electrostatics. One such prefilter used the electrostatic properties of electret fibers that carry a permanent electric charge and another employs a non-ionizing electric field in combination with a fibrous filter. Both developments are reported to give a spectacular improvement in filter efficiency with no increase in air flow resistance, either initially or as dust accumulates in the fiber structure.

Radioiodine collection

Preventing the release of volatile radioiodine (I*) is of major importance in the design of nuclear reactors because of the very low value (50 mcCi) that has been assigned for the maximum permissible body burden. This makes I* the single most important fission product to be considered when designing engineered safety systems for control of gaseous emission. The great concern over the presence of radioactive iodine in the atmosphere is brought out clearly by the enormous attention that was given to the release of 15 curies of I-131 from TMI-2 compared to the attention given to release of two and a half million curies of radiokrypton and Xe. Fortunately for emission control, most of the fission product iodine is elemental, a form that is easy to extract from an inert carrier gas stream, But a minor fraction, composed of organic and oxygenated iodine species, is less easily captured and these components have been the focus of I* removal efforts over the past two decades. The ability of chemical adsorbents, chemisorbents containing silver, and activated charcoal to extract iodine from waste gas streams was recognized then and all three were under vigorous investigation. Activated charcoal was often the preferred medium because of its unexcelled retention capability when fresh and dry. It was observed frequently that when activated charcoal beds were called into service they had become badly degraded from prior deposition of organic solvents and water vapor. Such degradation of charcoal has by now become such a widespread phenomenon that an inverse relationship has been noted between the life span of installed charcoal beds and the cleanliness of the facilities they serve; the shortest-lived beds resulting from the most frequent use of cleaning chemicals and paints.

Effects of moisture

By 1968, it was already well known that the disastrous effects of moisture on the efficiency of activated charcoal for organic iodides (principally methyl iodide) could be at least partially overcome by charcoal impregnation, either with inorganic iodides such as KI or KI_3 to provide I-131/I-127 isotopic exchange capability , or with highly reactive amines, such as hexamethylenetetramine (HMTA) and triethylenediamine (TEDA). At present, both impregnants are being added to the same nuclear grade charcoal; the preferred form of the inorganic exchange iodide often being potassium triodide (KI_3). But difficulties remain because charcoal degradation can occur as a result of the volatilization of the TEDA impregnant and from the deposition of water and organic solvents or other compounds that are held on the carbon tenaciously and block the deposition sites for methyl iodide. These effects are generally referred to as aging, weathering, and poisoning. Storage in sealed containers has little effect on charcoal; whereas weathering, i.e., passage of air through the charcoal containing water vapor and a variety of air contaminants such as NOx, SO, and O_3 can result in adsorption of water vapor, erosion of the charcoal surface, and volatilization of impregnants. Poisoning occurs when unintended components are picked up by the charcoal that prevent later adsorption and retention of methyl iodide. Remedial actions call for the development of organic impregnants that are less volatile than TEDA, yet retain equal or better chemical reactivity for organic iodides. One such compound receiving attention at this time is quinuclidine and, doubtless others are being sought.

Impregnated charcoal

Impregnated charcoal beds (F-6) intended for methyl iodide removal can be protected from poisoning by placing thin layers of unimpregpated, and hence far cheaper, charcoal upstream of the impregnated beds. Such an unimpregnated charcoal guard bed need not meet nuclear standards to be fully capable of removing all of the easily adsorbed compounds that can poison the nuclear-qualified impregnated charcoal bed and render it inefficient for methyl iodide. The guard bed can, of course, be changed out as often as needed to maintain its protective function. It is not clear to this observer why the use of guard beds has not yet become universal for nuclear installations concerned with I* retention. Perhaps recent events will hasten its adoption. Although the activated charcoal beds that became the primary iodine barrier at TMI were not included in the engineered safety systems for this reactor, the charcoal was nuclear grade and the installation had been leak tested when installed. Nevertheless, it became clear following the reactor accident that these beds had become seriously degraded through weathering and poisoning over the brief period of reactor service; one of the four installed banks retaining a mere 49% of the methyl iodide entering it.

Charcoal bed ignition

The combustible nature of activated charcoal troubled nuclear safety engineers from the beginning. It is well known that should a fire start in a charcoal bed, it can be extinguished only by heroic means, e.g., total and prolonged immersion in water or nitrogen. It was feared that if all the decay heat were to be concentrated in a thin layer inside the charcoal bed, it would be capable of initiating local combustion that could spread rapidly, and this sequence of events was readily demonstrated in the laboratory using adsorbed radioactive elements and stagnant beds of charcoal. It was also found that the presence of organic solvents on poisoned charcoal significantly lowered ignition temperature. This, incidentally, appears to be still another reason for insisting on the use of guard beds. Nevertheless, additional study has demonstrated that even a modest gas flow through the charcoal bed is more than sufficient to carry off all the decay heat that can be generated by adsorbed fission products. This has made it essential to provide for continuous gas flow through charcoal beds during and following the release of adsorbable radioactivity to prevent local overheating that would first, cause desorption of the more volatile components and later, cause ignition that would free the entire contents of the charcoal bed. This precaution has been incorporated into all engineered safety systems. As an additional precaution in the event of a charcoal fire, a deep bed of silver plated copper ribbon was installed downstream of the charcoal in the waste gas system of the Nuclear Ship Savannah. It was intended to function as an efficient chemisorbent when its temperature was elevated by hot gases from the burning charcoal and was expected to pick up all iodine desorbed from the charcoal.

Because fears of carbon ignition persist, there has been a continuing interest in non-combustible substitutes. A number of inorganic adsorbents have been investigated but silver-substituted zeolites have received the most attention because of their high efficiency for iodine and methyl iodide and their good iodine retention characteristics at temperatures up to 1000 C. However, there was considerable scepticism expressed about the practicality of silver-substituted zeolites, considering their cost, when the subject was discussed at the 1968 IAEA meeting. More recently this objection has been countered by a plan to reactivate saturated silver-substituted zeolites by transferring the adsorbed I* to lead-substituted zeolites for permanent disposal by long term storage, but the current price of silver on the international market does not make the use of silver-substituted zeolites attractive for any purposes for which a reasonable substitute is available and it is unlikely that silver will receive serious consideration as a universal substitute for activated charcoal until the cost of silver declines. A significant development in charcoal adsorption technology for the capture of I* has been the introduction of dumped, deep beds of charcoal, 6 or more inches thick, often called gasket less charcoal beds, as a substitute for prepackaged trays or cells containing a 2 in. thickness of tightly-packed charcoal. The Reactor Safety Commission in Germany now requires a 20 cm thickness of charcoal and is considering increasing depth to 50 cm. These deep beds can be provided with automatic, remotely controlled mechanical means to change the charcoal filling, thereby eliminating human exposure to the collected radiation. The thickness of the charcoal in the direction of airflow compensates for an inability to vibrate these beds to achieve maximum packing density of the granules and provision is made to overfill the beds so that in the event of settling, there will always be an excess of charcoal to fill the voids. Nevertheless, it has proven very difficult to locate and eliminate leaks in these deep beds when revealed by Freon leak testing and often it is necessary to withdraw the charcoal completely and refill the bed repeatedly before a satisfactory fill can be achieved. The principal difficulty associated with these beds, however, is an inability to withdraw a representative sample of charcoal from the bulk filling for the required periodic residual life tests.

Krypton and xenon collection (IAEA 1980)

Large amounts of noble gas radioactivity have been vented to the atmosphere since the beginning of the atomic age and it continues to the present time. Because these gases have little biological activity and most have a short half-life, they have not been considered a serious problem up to the present; although it is recognized that ever-increasing discharges of these radioactive gases to the atmosphere may result, eventually, in an excessive worldwide external radiation dose to populations. Three distinct systems have been developed for removing noble gases from waste gas streams: absorption in cooled fluorocarbon liquids, low temperature adsorption on activated charcoal at elevated pressure and cryogenic separation. Cryogenic separation has become the method of choice at fuel reprocessing plants in Germany, and perhaps elsewhere in Europe. A low temperature - high pressure charcoal adsorption unit has been installed at the Fast Flux Test Facility at Hanford, Washington, to free the helium cover gases of Kr and Xe. Fluorocarbon absorption has failed to attract users and, for the moment at least, is in eclipse, leaving the entire field to charcoal and cryogenic separation. All of the separation systems provide a means of concentrating Kr-85 as a preparatory step for extraction, compression, and long storage in pressure cylinders. Each of the three methods that have been developed to separate noble gases requires a considerable capital investment in equipment and substantial operating costs for temperature and pressure regulation. As a consequence, they can be applied most efficiently to concentrated waste gas streams such as those that result from spent fuel processing. To date, noble gas treatment of gaseous effluents from light water reactors has generally consisted of brief holdup in charcoal beds operated at atmospheric temperature and pressure to permit decay of those isotopes having a short half-life, e.g., holdup of 1-3 days for Kr, 10-25 days for Xe. Up to the present, the events at TMI-2 have failed to generate strong opinions for change but should it become desirable to reduce emissions of noble gas activity to the atmosphere from power reactors, there appears to be a satisfactory technological basis for removing them from all waste gas streams that contain sufficient radioactivity to make extraction worthwhile.

Tritium removal (IAEA 1980)

Gaseous releases of tritium from light water reactors have not been a serious problem but the increased emphasis on minimizing radioactive effluents, the ALARA principle (as low as reasonably achievable) initiated by the USAEC and continued by NRC, has stimulated interest in removing even small amounts prior to the discharge of waste gases. The methods that may be used for removing tritium involve straightforward chemical reactions that involve catalytic oxidation of uncombined tritium, cooling to condense tritiated water or to adsorb it on molecular sieves, and reduction back to elemental form to produce a highly concentrated tritium stream for storage in compressed gas cylinders. Numerous variations of this basic treatment process are employed by operators of heavy water cooled reactors and fuel reprocessing plants, and interest in the recovery and recycling of heavy hydrogen is associated with the fusion energy program. For all these applications, the technological basis for removal of tritium from waste gas streams seems to be sufficiently well developed to be satisfactory for the years ahead.

Carbon-14 removal

The technology available for extracting C-14 from waste gas streams also appears to be adequate for the years ahead. In general, it, like tritium extraction, involves straightforward inorganic chemical reactions, e.g., oxidation of all carbon compounds to carbon dioxide and extraction of the resulting CO_2 in reactive solutions or on solid chemisorbents from which it can be desorbed in concentrated form for final disposal.

Discharge of radioactive effluent to the atmosphere (Collins 1960)

Many operations in factories which use or process radioactive materials result in radioactive airborne or gaseous effluent. Such effluent may be entirely gaseous, entirely particulate or, as often happens, it may be present partly in the vapour phase and partly adsorbed on small particles. The volumes of gas involved are often very large and the discharge of the effluent to atmosphere may at first sight appear to be an attractive method of disposal. However, the effect of incautious releases to atmosphere is to leave the operator to the mercy of the four winds for, while being deprived of control, he is by no means relieved of responsibility. The necessity of treating large volumes of gas so that their release to atmosphere will not cause a hazard, either to the employees of the factory or to the general public, has given rise to many novel problems. The assessment of these problems and the solutions which have been arrived at for some of them will be discussed in this chapter. The emission of activity to the atmosphere may give rise to three possible types of hazard - a direct irradiation hazard from the radioactive cloud itself or from material which is deposited on the ground, an inhalation hazard to people breathing the cloud and an ingestion hazard from material which finds its way into food chains. The type of hazard which will predominate depends on the circumstances of a particular emission. For alpha emitting isotopes, for example, the predominating hazard will invariably be from ingestion of material either inhaled directly from the cloud, or initially deposited on the ground and subsequently blown up again by the wind, or eaten in food. For most other isotopes the predominant hazard will be caused either by direct irradiation from material deposited on the ground or by ingestion of such material with food.

Behaviour of effluent plumes

Consider an emission of radioactivity from a stack. As the gas is discharged from the stack, it will carry with it all particulate matter whose terminal velocity is less than the stack gas velocity. On emerging from the stack the effluent becomes subject to the wind and suffers a change in direction and a diminution of upward velocity. The larger particles begin to fall out and, as the cloud drifts further away from the stack, smaller and smaller particles fall to the ground. In the meantime the cloud itself is growing larger and more dilute as a result of turbulent diffusion in the atmosphere and eventually its dimensions are such that it sweeps along the ground. At this stage deposition by impaction and turbulent diffusion occurs of vapours in the cloud and of those particles which have such a small terminal velocity that they have not previously fallen out under gravity. The behaviour of the cloud for a given set of conditions, and the extent of dilution and deposition at a given distance from the site, are the factors which determine the hazard which can arise. They will now be studied in some detail.

Release from a stack

Effluent emitted from a stack at first continues to rise because of its exit velocity and any buoyancy which it may possess by virtue of an excess of temperature over that of the atmosphere. The subsequent behaviour of the plume is thus as if the effluent had been emitted from a height greater than the stack height. It was found that for gas at 50 C above atmospheric temperature emerging at 10 m/s from a stack 61 m high, the effective height of the plume was about 125 m. Numerous attempts have been made by various authors to derive a formula by which the 'effective height of release' or 'equivalent stack height' may be calculated for a particular stack. One of the simplest of these formulae is the Oak Ridge formula:

$$Z = (1.5vd + 0.4Q)/u \qquad (4)$$

where Z = height reached by effluent above top of stack (m); u = wind velocity (m/s); v = exit velocity of effluent (m/s); d = chimney diameter (m); Q = heat output (calories/s).

It is a matter of common observation that the smoke plume from a particular factory chimney may rise to different heights from day to day and even at different times on the same day. All such formulae are limited in their usefulness by the fact that they cannot hope to take account of the wide variation of meteorological conditions which may occur. In particular, the Oak Ridge formula gives the effective height of the plume from the Bepo reactor of the United Kingdom Atomic Energy Authority as 76 metres compared with the 125 metres determined experimentally.

The existence of temperature inversions at various levels may also play havoc with the applicability of theoretical formulae. If the stack penetrates an inversion layer, the subsequent diffusion of the plume to ground level will be inhibited and the effective height of release will be infinite. This is, in practice, one of the greatest advantages of tall stacks. However, the converse is not necessarily true and the fact that it is possible, even when an inversion extends above the top of a stack, for hot effluent to pass through the inversion layer, has been demonstrated in London by the Central Electricity Generating Board. In conditions of thick fog, the plumes from several power stations, coloured red and black for the occasion, were observed from an aircraft to penetrate the top of the fog layer and disperse in the upper atmosphere.

In addition to meteorological factors, the rise of the plume may be affected by the physical surroundings of the stack. Eddies caused by neighbouring tall buildings may draw the effluent prematurely to ground level and a useful empirical rule is that the stack height should be at least two and a half times that of neighbouring buildings. A further problem is that of 'downwash', which usually occurs when the exit velocity of the effluent is low. Instead of rising from the stack, the plume spills over the leeward edge and creeps down the outside wall. This phenomenon has been known to cause erosion of concrete stacks and is clearly even less desirable with radioactive effluent. The remedy is to increase the exit velocity of the effluent to the same order as the wind velocity. In a comprehensive study of the behaviour of chimney plumes, Scorer gives an alternative solution of installing at the top of the stack a horizontal ring extending outwards from the stack to a distance of one stack diameter.

Diffusion in the atmosphere

The extent of diffusion in a particular air mass is strongly influenced by the variation of air temperature with height. In the particular case where the thermal structure of the atmosphere is such that a parcel of air, when it is displaced from one level to another, always retains the same density as the surrounding air, the rate of decrease of temperature with height, or lapse rate, is known as the 'dry adiabatic lapse rate', and is numerically equal to a decrease of temperature of 1 C per 100 metres. If atmospheric temperature decreases with height at a rate greater than the dry adiabatic, a parcel of air, on being displaced upwards adiabatically, would be at a higher temperature and of less density than the surrounding air and hence it would continue to rise. Similarly, if the air was displaced downwards adiabatically, it would be at a lower temperature and hence be more dense than its surroundings and would tend to sink further. Thus for lapse rates greater than adiabatic, vertical motions are accelerated and the atmosphere is said to be unstable. On the other hand, when the lapse rate is lower than adiabatic, a parcel of air displaced upwards will have a greater density than the surrounding air and will tend to return to its original position. In such conditions the atmosphere is stable and, when the lapse rate is positive, (that is to say temperature increases with height), an inversion is said to obtain, atmospheric conditions are extremely stable and vertical diffusion is minimal.

The effect of these various temperature gradients on the diffusion of effluent emerging from a stack is as one would expect from the foregoing discussion. Strong lapse rates favour rapid diffusion and dilution, while vertical diffusion in inversion conditions is extremely slow. Meteorological conditions which are a combination of the two often arise. For example, inversion conditions may occur below a certain height and lapse above. In this instance, if a stack penetrates the inversion layer, diffusion of effluent gas to the ground will tend to be inhibited. On the other hand, if lapse conditions occur up to the height of the plume and inversion above, upward diffusion will be reduced and diffusion to ground level enhanced.

Treatment by Sutton's equations

Considerable effort was applied in the interwar years to the theoretical treatment of the problems of the diffusion of smoke and gas sources in the atmosphere. It is not proposed to discuss the various theories which have been evolved but to quote from one of them, that due to Sutton, which has been widely applied and checked experimentally in certain meteorological conditions. Sutton derived the following expression for the concentration from a ground level point source:

$$K(x,y,z) = 0.64qx^{n-2}*\exp(-y^2C_y^{-2}x^{n-2} - z^2C_z^{-2}x^{n-2})/u/C_y/C_z \qquad (5)$$

where $K(x,y,z)$ = concentration (units/m^3) at a point x metres downwind, y metres crosswind and at height z metres; q = rate of emission of ground level point source (units/s) ; u = wind velocity (m/s); C_y = Sutton's virtual diffusion coefficient in the cross-wind direction ($m^{1/8}$); C_z = Sutton's virtual diffusion coefficient in the vertical direction ($m^{1/8}$); n = constant depending on the lapse rate (dimensionless).

Table 18 Constants applicable to Sutton's equations.

Atmospheric condition	n	C_y	C_x
Large lapse rate	0.2	0.37	0.21
Zero temperature gradient	0.25	0.21	0.12
Moderate inversion	0.33	0.13	0.08

It will be noted from equation (5) that the dimensions of C_y and C_x depend on the value of n and for n = 0.25, C_x and C_y have dimensions in $m^{1/8}$. From this equation the expected result is obtained that, for a given distance x from the source, the maximum concentration occurs directly downwind of the source at ground level, and is given by:

$$K = 0.64qx^{n-2}/C_z/C_y \qquad (6)$$

and by integrating both sides of equation (6) with respect to time, the time integral of concentration, or dosage, is obtained for a source which has been released in a relatively short time:

$$D = 0.64Qx^{n-2}/C_z/C_y \qquad (7)$$

where D = dosage (units-seconds/m^3); Q = total release (units).

Curves of K/q, the dilution factor, against x (for a wind speed of 5 m/s) are given for the three sets of meteorological conditions. At a given distance from the source there is roughly an order of magnitude increase in the values of the dilution factor as conditions go from large lapse to moderate inversion.

Release at a height

Sutton extended his theory to cover elevated sources. He assumed that the same value of n would apply and derived theoretically values of C_z and C_y which varied with height of release. The equation for the concentration at ground level directly downwind of an elevated source is:

$$K = 0.64qx^{n-2}*\exp(-h^2C_z^{-2}x^{n-2})/u/C_z/C_y \qquad (8)$$

where h = height of release (metres).

For h = 100 metres, Sutton gives C_z = C_y = 0.07, but experimental evidence is that better agreement is obtained with the theory if the values of C_z and C_y applicable to ground level release are used. From a study of sulphur dioxide pollution around Staythorpe power station, Meade and Pasquill deduced values of C_z of 0.21 m$^{1/8}$ (winter) and 0.35 to 0.42 m$^{1/8}$ (summer) at a distance of 1370 m. Stewart suggests values at a distance of 800 metres of 0.11 m$^{1/8}$ (inversion conditions), 0.20 m$^{1/8}$ (adiabatic conditions) and 0.25 m$^{1/8}$ (lapse conditions) and at distances up to 1050 metres a value of 0.32 m$^{1/8}$. To eliminate the effect of different wind speeds in the various experiments, the dilution factor is tabulated in the form Ku/q and the values given for this function are at the stated distance directly downwind from the stack. The fact that different experimenters used different sampling times introduces a further complication, since the highest concentrations may only be present for a very short time, and the longer the sampling time the greater the tendency for the peaks to be flattened out. The value of h used in equation (8) to obtain the measured values is the height of the stack concerned and no allowance has been made for the additional height reached by the effluent plume because of its exit velocity and temperature. The agreement between the experimental value of the dilution factor and those calculated from Sutton's equation with C_y = 0.21 and C_z = 0.12 or 0.21 is remarkably good, most of the experimental values lying within a factor of two of the theoretical (up to distances of 10 000 metres from the source). The applicability of Sutton's theory in inversion conditions is unfortunately not so well established.

At a distance of 20 effective heights of release from the source, the ground level concentration due to release from a height of 10 metres is 80 % of that due to an equivalent ground level release. For release at a height of 100 metres, the figure is 68%. It thus appears that reduction in the ground level concentration obtained by release from a stack is greatest up to a horizontal distance from the source of about 20 times the effective height of release. Beyond this distance, in average meteorological conditions ground level concentrations are little different from those due to an equivalent ground level release.

Deposition

To describe the deposition behaviour of aerosols, the concept of velocity of deposition, v, was introduced :

v = total deposition per unit area/time integral of volumetric concentration

For large particles, v is equal to the terminal velocity, but an aerosol of negligible gravitational settling velocity may have a high velocity of deposition by diffusion and impaction. For larger values of v the rate of deposition relative to the volumetric concentration at a point is increased, but the concentration of the cloud at that point has already been reduced, because of the relatively greater deposition which has already occurred. It follows that for a given distance from a source there is a certain value of v which will give the maximum possible deposition at that distance. Chamberlain modified Sutton's equation to allow for deposition and obtained the rate of deposition at point (x,o) due to a ground level release:

$$w(x,o) = 0.64qvx^{n-2}*\exp(-2.26vx^{n/2}/n/u/C_x)/u/C_y/C_z \qquad (9)$$

The maximising value of v, which will give maximum deposition at distance K, is obtained by differentiating (9) with respect to v, and equating to zero, whence:

$$v* = 0.44nux^{-n/2}C_z \qquad (10)$$

and for $n = 0.25$, $C_z = 0.21$ and $u = 5$ m/s, this reduces to:

$$v* = 0.116x^{-0.125} \qquad (11)$$

and gives values of v* of 5 cm/s at a distance of 800 metres and 2 cm/s at 1E+6 metres.

Some experimental values of the velocity of deposition (in cm/s) of various aerosols obtained on the grass airfield at Harwell, England, are : lycopodium spores (32 microns) 1.2, I-131 vapour 2.5, fission product aerosol 0.1 and strontium and barium fractions of fission product aerosol 0.01. It is seen that there is a variation between them amounting to a factor of 250. The modes of deposition of the four aerosols were very different. The lycopodium spores were deposited primarily by virtue of their terminal velocity (2 cm/s), and the iodine vapour by diffusion. The fission product aerosol was generated by striking an electric are between irradiated uranium electrodes and that its terminal velocity was very small was indicated by the fact that deposition on upward and downward facing surfaces was equal. It is possible that interception by grass leaves, diffusion and electrostatic deposition all contributed. The velocity of deposition of the strontium and barium fractions of the fission product aerosol was determined separately by means of radiochemical analyses of the samples and found to be less by a factor of 10 than for the mixed fission products.

Adsorption on condensation nuclei

There are always present in the atmosphere numbers of small particles, Aitken nuclei, of diameter a few hundredths of a micron. They are formed in large numbers by most combustion processes and are present in concentrations which may vary from a few thousand per ml in clean country air to more than a hundred thousand per ml in urban areas. It has been known for some time that most of the radon and thoron decay products present in the atmosphere are attached to condensation nuclei. It was shown that the deposition of such decay products is reduced by a factor of about 500 by being attached to Aitken nuclei which are of such a size that, while having a negligible settlement rate (about 1E-4 cm/s), they also have a low diffusion constant (about 1E-5 cm^2/s) compared to that of the unattached decay product atoms. The velocity of deposition of the fission products present in the atmosphere as a result of nuclear weapons tests is equal to 0.014 cm/s for deposition on filter paper. There appears to be at any rate a strong inference that the reason for the low velocity of deposition found for strontium and barium was that the strontium and barium atoms had become attached to Aitken nuclei before deposition. The progressive reduction, with increasing distance from the source, of the velocity of deposition of the iodine released in the Windscale incident suggests that a proportion of the iodine was adsorbed on nuclei. The presence of condensation nuclei may greatly reduce the deposition of radioactive effluent and it is possible that future developments may enable practical use to be made of the effect.

Washout by rain

The ground deposition deduced above may be increased by rain falling through the effluent cloud. The theory was confirmed by experimental work using an aerosol of lycopodium spores (diameter 32 microns). From the theory, for a given aerosol and rate of rainfall it is possible to calculate the proportion of the effluent cloud which is washed out per second. For example, if the cloud consists of 10 micron diameter particles of density 2.5 and rain is falling at the rate of 1 mm/h, 1 part in 3000 of the cloud is washed out per second and half is deposited in 36 minutes. Theoretically, the worst possible deposition by rain is 8 times the worst possible deposition in dry weather. It is common experience that most of the fallout from nuclear weapons tests is in rainfall and in the area South-East of Windscale Works, Cumberland, in which I-131 was deposited as a result of the incident in October, 1957, there was a narrow band of increased deposition which corresponded to the direction of the line joining the Pile 1 stack to the cooling towers of Calder Works. It seems probable that this increased deposition was caused by washout from the cloud by condensed moisture from the cooling towers.