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An aging line especially designed for the aging of pollutant atmospheres is described. The line essentially is a nearly-square stainless steel duct with periodic 100-mesh stainless steel screens to promote plug flow. The line has been tested for uniformity of aging, aerosol losses, and ease of atmosphere control. Results show that a close approximation to plug flow can be achieved with excellent atmosphere control; aerosol losses in the particle size range 0.22-2.0 μm are less than 30% except at very long (96 min.) aging times, when significant sedimentation losses occur for larger particles.

An aerosol and gas aging line suitable for use in inhalation toxicology research

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introduction

Inhalation exposures to air contaminants occur only after transport, and hence aging, of the original emissions. During aging many physical and chemical processes occur that may significantly alter the properties, and hence toxicity, of the original emissions. Such processes include: dilution, gas-to-particle conversion, adsorption, desorption, coagulation, charge equilibration, chemical reactions, and radioactive decay.

For the purpose of clarity, the aging time is defined as the elapsed time after components of a mixture are combined. The mixture may be batch generated, as in a spill or explosion, or continuously generated, as in a smoke stack. Air-

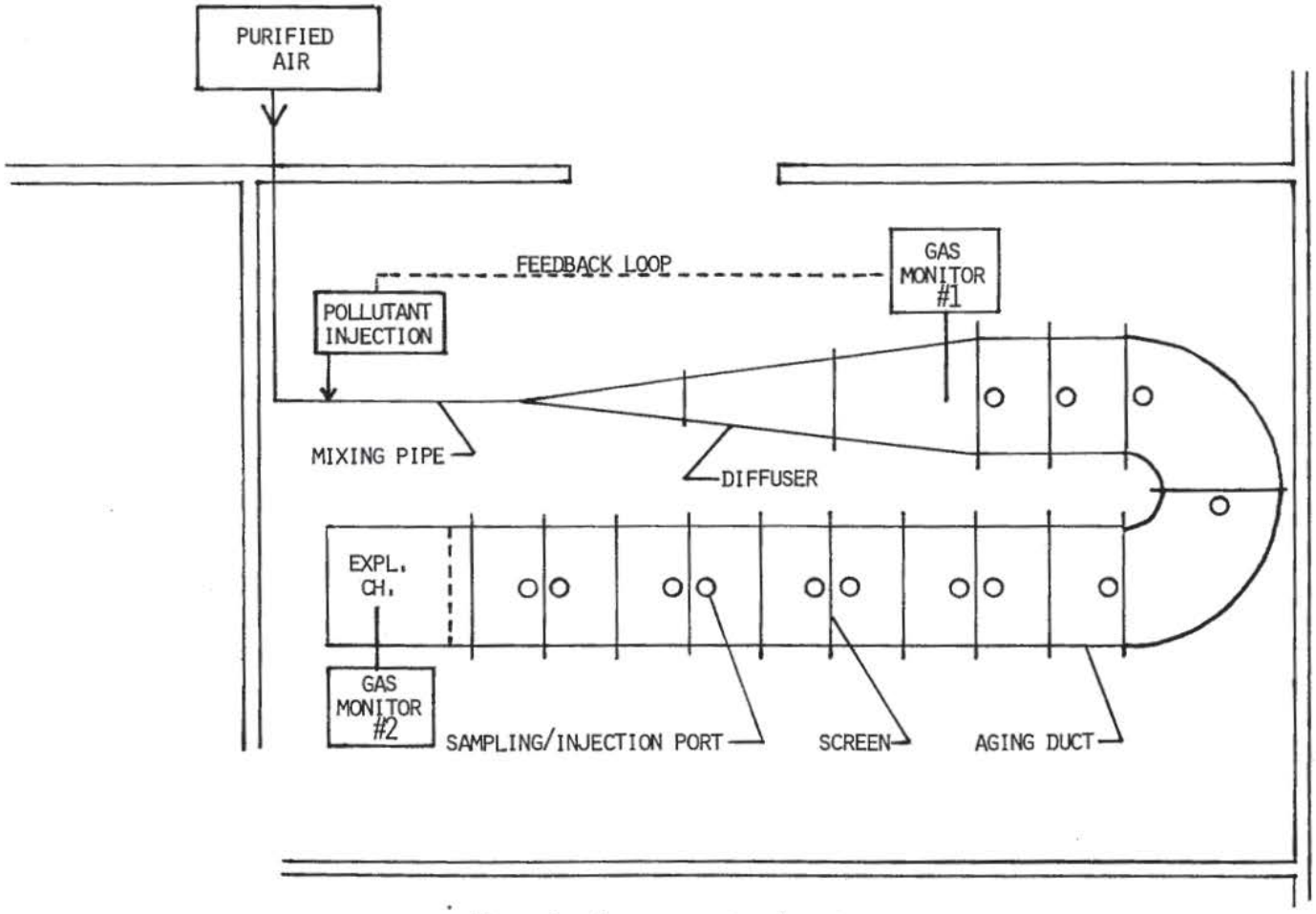

Figure 1 — Exposure system layout.

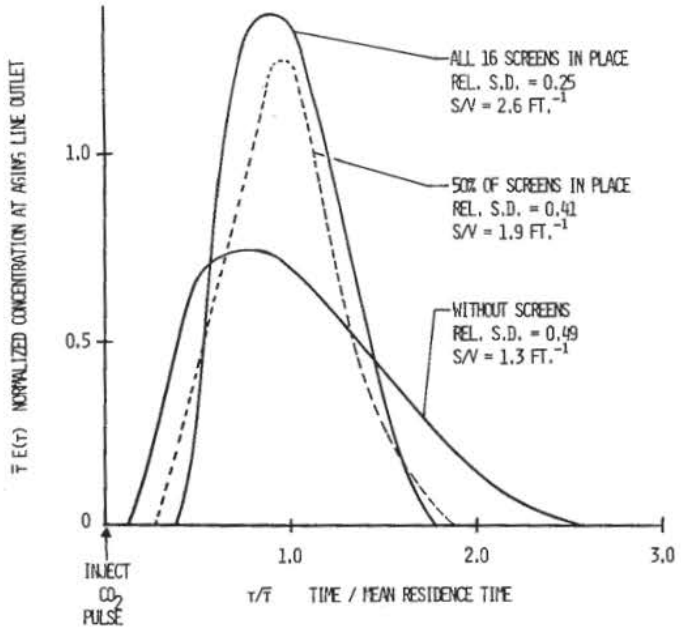

Figure 2 — Effect of screen addition on carbon dioxide gas residence time distribution in aging line (mean residence time 30 minutes).

borne mixtures may reach the breathing zones of people after aging times ranging from seconds, as in the case of cigarette smoke and industrial plant contaminants, to hours or days, as in the case of outdoor air pollution plumes.

There are two basic approaches one may take when investigating the toxicity of a mixture that changes with aging. First, if the physical and chemical properties of the atmosphere are well understood, with the kinetics and identities of all major chemical intermediates and end products known, then the chemical composition of the mixture at any aging time will be known and the mixture can potentially be generated. When direct generation of intermediates and endproducts is practical, such an atmosphere could be studied at any aging time in a conventional non-delay exposure system. Often, however, the kinetics of complex pollutant mixtures are poorly understood, with little knowledge as to type and quantities of reactive intermediates present. A case in point are the mixed atmospheres containing ozone, sulfur dioxide, ammonium sulfate, and ferric sulfate which are currently under study at this laboratory in an attempt to simulate mixed oxidant and sulfur plumes of the type found downwind from power plants. The expected aging time of these mixtures in the atmosphere before possible inhalation will include times considerably greater than the typical 1-10 minute delay time of conventional inhalation chamber exposure systems.

These considerations prompted the development of a gas/aerosol aging line with an aging capability of roughly an order of magnitude greater than conventional exposure chamber systems. If toxic effects are seen only after significant aging times it is likely that the toxic factors will not be identified until the chemistry of the system is understood. However, by selective subtraction of reactants until toxic effects disappear information immediately valuable to the planning of emission control strategy will be gained.

design considerations

A number of factors should be taken into consideration prior to the design of any system for the aging of reactive atmospheres. Early in the design process, a decision must be made between static and dynamic systems. In a static system air does not flow past the exposed subjects. Dynamic flowing systems are generally preferred for whole-body exposures of animals due to the increased ease of maintaining constant temporal and spatial concentrations of the pollutant atmosphere and the minimization of animal byproduct buildup problems. Aging times must be sufficient for all of the desired physical or chemical transformations of the experimental atmosphere to take place. Uniformity of aging time is important in many circumstances; this necessitates a close approach to plug-type flow in the aging system. Aerosol and gas losses in the system should be held to a minimum to reduce the frequency of system cleaning and to facilitate atmosphere control.

description of the apparatus

A plan view of the aging system is presented in Figure 1. Pollutants are continuously injected into 85 to 850 Lpm purified air in a turbulent mixing pipe connected through a diffuser to the aging line, an 89 cm high by 99 cm wide horizontal rectangular stainless steel duct 9.3 m long.

Flow through the duct is always laminar with the Reynolds number based on the hydraulic duct diameter

$$\mathrm{Re} = \frac{2\,V\,w\,h}{(w+h)\,\nu} \qquad (1)$$

ranging from 100 to 1000 depending on aging line flowrate. In this equation, V is the superficial duct air velocity, w and h width and height of the duct, and ν the kinematic viscosity of

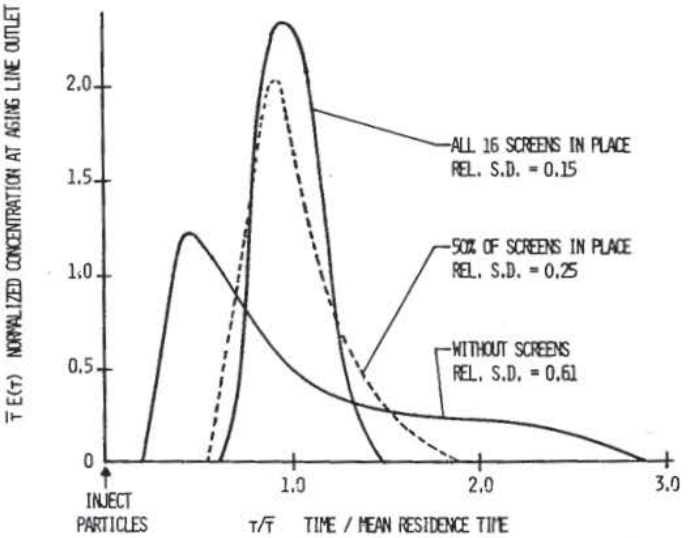

Figure 3 — Effect of screen addition on ammonium sulfate particle residence time distribution in aging line.

TABLE I
Aerosol Losses in Aging Line at 96 Minute Mean Aging Time

PSL Particle Diameter (μ)	Measured Loss OPC (%)	Measured Loss EM (%)	Theoretical Loss Sed. Loss (%)	Theoretical Loss Screen Loss (%)	Theoretical Loss Total Loss (%)
0.22	*	20	2	38	40
0.46	11	0	6	23	29
0.95	28	18	21	14	35
2.02	62	61	85	12	97

*Particle Size Below Instrument Range

TABLE II
Aerosol Losses in Aging Line at 10 Minute Mean Aging Time

PSL Particle Diameter (μ)	Measured Loss OPC (%)	Theoretical Loss Sed. Loss (%)	Theoretical Loss Screen Loss (%)	Theoretical Loss Total Loss (%)
0.22	*	0	10	10
0.46	8	1	6	7
0.95	10	2	4	6
2.02	12	8	4	12

*Particle Size Below Instrument Range

air. Removable 100-mesh stainless steel screens are mounted at 60 cm intervals along the duct to flatten the air velocity profile and promote plug flow; 5 cm diameter ports are welded along the length of the duct for sampling and cleaning purposes. The outlet of the aging duct is directly connected to an exposure chamber through a large elbow.

experimental methods

The completed system was tested for uniformity of aging, aerosol losses, and ease of atmosphere control.

In order to obtain information on the mixing characteristics of the aging line, a population balance model was employed.[1] A given vessel of volume B is assumed to contain N elements of volume b, such that $B = Nb$. Although b/B is small, the fluid elements are assumed to contain sufficient molecules so that macroscopic properties such as reactant concentration and fluid density can be defined. The age of a fluid element at any given time is the time elapsed since the element entered the vessel; since all the fluid elements leaving the vessel must have some age, it follows that

$$\int_0^\infty E(t)\,dt = 1 \qquad (2)$$

where E(t) is the fraction of fluid elements in the vessel exit stream with ages between t and (t+dt), *i.e.* the fluid element residence time distribution function for the vessel.

A stimulus/response tracer injection technique was used to measure this distribution function and thus characterize the extent of nonideal flow in the line. A pulse of tracer (CO_2 gas or 0.2 μm MMD $(NH_4)_2SO_4$ aerosol) was injected into the aging line inlet, and concentrations of gas or aerosol were measured at the aging line outlet (animal zone of exposure chamber) using a medical mass spectrometer (Perkin-Elmer Model 1100) or condensation nuclei counter (Environment-One Model Rich 100), respectively. Periodic measurements were continued until the fresh air inflow to the aging line had completely washed the tracer from the system and tracer material could no longer be detected. The tracer experiments were conducted with zero, 8, and 16 100-mesh stainless steel screens installed at approximately equal intervals along the aging line.

Aerosol losses in the system were measured at 10% R.H. by nebulizing a dilute aqueous mixed suspension consisting of equal concentrations of 0.22, 0.46, 0.95, and 2.02 μm diameter polystyrene latex spheres (Dow Chemical Co.). The nebulizer output was dried, discharged, and injected into the mixing pipe; the resulting aerosol contained less than 5% doublets. Aerosol concentrations after mixing were low enough to make coagulation effects insignificant. After attainment of equilibrium, aerosol samples were taken at the diffuser outlet and animal zone of the exposure chamber using an optical particle counter (Climet Model 208) and point-to-plane electrostatic precipitator (ARIES, Inc., Davis, CA). The ESP grids were shadowed with platinum and analyzed by transmission electron microscopy.

Degree of atmosphere control was evaluated by generating a 2 mg/m^3 internally mixed ammonium sulfate/ferric sulfate aerosol in combination with 0.8 ppm ozone and 5 ppm sulfur dioxide at 85% relative humidity, and monitoring the time stability of the gas concentrations in the animal zone of the exposure chamber for a 4-hour period after equilibration. An ultraviolet absorption-type O_3 monitor (Dasibi Model 1003AH) and a pulsed-fluorescent SO_2 analyzer (Thermo-Electron Model 43) were used for the gas measurements.

results and discussion

The measured residence time distribution functions for gas and particulate tracers, with and without screens, are shown in Figures 2 and 3. The data points were used to draw empirical curves shown in the figures. As the number of screens is increased, a pronounced decrease in peak relative standard deviation occurs for both gaseous and particulate tracers indicating a flow transition from perfectly mixed behavior (relative standard deviation = 1.0) towards ideal plug flow (relative standard deviation = 0).

The remaining deviation from ideal plug flow may be due to tracer diffusion effects and the effect of lateral flows. At the lower aging line flowrates, superficial velocities are the same order of magnitude as thermally-induced convection currents resulting from temperature gradients. Further, secondary flows will appear in the boundary-layer development regions between screens and in the curved portions of the aging line since the Dean number

$$D = \frac{V\, d^{1.5}}{\nu\, R_1^{.5}} \qquad (3)$$

in the aging line is greater than the critical value of 36.[2,3] Here d is the spacing between the inner and outer walls and R_1 the radius of curvature of the inner wall of the elbow; the Dean number is proportional to the ratio of centrifugal forces to viscous forces acting on the fluid.

The use of screens as flow redistributors is well known and has been recently reviewed by Laws and Livesey.[4] The pressure drop across a screen is given by

$$\Delta p = \tfrac{1}{2} K \rho V^2 \qquad (4)$$

where ρ is the air density and K the screen resistance coefficient. A minimum K value of 2.8 is required to produce downstream uniformity for any upstream profile, and for screen Reynolds numbers below 250 (the usual case) with incompressible flow K depends only on screen porosity (ratio of open to total screen area).[4] In the present design, duct air velocities are kept low enough so that the static pressure drop across the entire aging line with all screens in place is less than 0.3 cm water column.

In addition to higher pressure drop, introduction of screens presents other possible disadvantages. As shown in Figure 2, the surface-to-volume ratio of the aging line was doubled by addition of sufficient screens to create near-plug flow. This characteristic may be objectionable when studying atmospheres where wall effects are significant. Secondly, aerosol losses in the aging duct will be increased. Under normal aging line operating conditions, aerosol sizes range from 0.1 to 2.0 μm while superficial duct velocities are 1.7 cm/sec or less. Under these conditions, the Stokes number

$$St = \frac{2\, \rho_p\, C\, r_p^2\, V}{9\, \mu\, r_w} \qquad (5)$$

and interception parameter

$$I = \frac{r_p}{r_w} \qquad (6)$$

are both very small, indicating that diffusion will be the dominant collection mechanism for 100-mesh screens. Here, ρ_p is the particle density, C the Cunningham slip correction, r_p the particle radius, μ the absolute air viscosity, and r_w the screen wire radius. The introduction of turbulence by the screens under the operating conditions stated above does not occur since screen Reynolds numbers are in the creeping motion region ($Re < 1$) and thus well below the range at which boundary-layer separation and vortex shedding are known to occur.[5]

The results of the aerosol loss study are presented in Tables I and II. In these experiments, losses were measured with all screens in place in the aging line at flowrates of 85 and 850 Lpm. These flowrates are close to the minimum and maximum practical flows through the aging system. Fine particles are lost primarily by diffusion to the screens, while sedimentation is the dominant loss mechanism for larger particles. The right-hand columns of the tables present theoretical expected losses calculated from the fan model filtration theory[6] and the gravitational settling equation

TABLE III
Atmosphere Stability Results

Laboratory	Reference	Length of Exposure (hrs.)	$\bar{x}$ (ppm)	sd (ppm)	sd/$\bar{x}$	$\bar{x}$ (ppm)	sd (ppm)	sd/$\bar{x}$	Atmosphere Studied
This		4	0.81	0.04	0.05	5.0	0.2	0.04	0.8 ppm O_3 +
Laboratory		4	0.80	0.04	0.05	5.1	0.1	0.02	5.0 ppm SO_2 +
		4	0.79	0.04	0.05	5.0	0.1	0.02	1 mg/m^3 $(NH_4)_2SO_4$ +
		4	0.82	0.03	0.04	5.0	0.1	0.02	1 mg/m^3 $Fe_2(SO_4)_3$,
		4	0.80	0.02	0.03	5.0	0.1	0.02	aged for 30 minutes
		4	0.82	0.02	0.02	5.0	0.1	0.02	
Laboratory A	(8)	2	0.36	0.02	0.06	0.36	0.02	0.06	$O_3 + SO_2 + 0.1$ mg/m^3 H_2SO_4
Laboratory B	(9)	*	0.80	0.08	0.10				O_3 only
Laboratory C	(10)	3	1.0	0.10	0.10				O_3 only
Laboratory D	(11)	2	0.48	0.07	0.15				O_3 only

*Intermittant Exposure

$$F = \frac{V_s L}{Z V} \quad (7)$$

where F is the fraction of particles lost, V_s the settling velocity of particles, L the aging duct length, and Z the duct dimension parallel to gravity.

The areal density of particles of a given size in electron micrographs of randomly-selected areas on ESP grids was assumed proportional to the airborne number concentration of particles of that size. To assure a constant coefficient of proportionality, care was taken to standardize sampling conditions. Even so, due to probable temporal fluctuations in ESP efficiency and non-uniform grid deposition, the ESP-derived results can be considered approximate only. With this limitation in mind, the agreement between the OPC and EM methods is satisfactory. As shown in Table I, experimental particle losses were consistently lower than losses predicted from theory at the lower aging line flowrate; the difference may be due to aerosol resuspension effects in the duct. Clearly, however, this system is not suitable for 96-minute aging of aerosols larger than a few microns in size. Agreement between theory and experiment at the higher flowrate was satisfactory.

The aging line has been used for acute 4-hour whole-body exposures of rats to mixtures of 2 mg/m^3 ammonium and ferric sulfate + 0.8 ppm O_3 + 5 ppm SO_2, aged for 30 minutes. With this long aging period it was initially felt that atmosphere control problems would result due to the long time lag. However, by inserting an extra SO_2 and O_3 monitor at the aging line inlet (Figure 1) and using this concentration information in a manual feedback control system to the gas generators, excellent control performance has been attained. For 0.8 ppm O_3 + 5 ppm SO_2 + 2 mg/m^3 $(NH_4)_2SO_4$/ $Fe_2(SO_4)_3$ atmospheres aged for 30 minutes, the required setpoint concentration for O_3 at the aging line inlet is slightly higher (0.95 ppm O_3) due to ozone decomposition losses in the aging line. For SO_2, setpoint and target concentrations are essentially the same under these conditions. No inlet concentration monitoring is done for the aerosol component, satisfactory time stability being obtained by controlling metering rate of aerosol into the aging system.

Table III presents gas concentration stability data for all exposures performed to date with the aging system; the control performance compares favorably with non-aged atmosphere stability data recently reported in the literature.

conclusions

A gas/aerosol aging line has been designed, tested, and successfully used for acute whole-body exposures of rats to atmospheres simulating urban-type mixed oxidant and sulfur plumes after 30 minutes of aging. Although the design is appropriate for the present application, further refinements could be added depending on the needs of the experimenter. These include:

1. Provision for rotation of the aging duct to reduce sedimentation losses for larger particles.[7]
2. Addition of photochemical capability.
3. Optimization of screen mesh size and porosity to minimize diffusion losses of fine particles.
4. Provision for longer aging times.
5. Electropolishing of inner surfaces to reduce micropore surface area and improve cleanability.

acknowledgements

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