Tracer-Gas Mixing with Air: Effect of Tracer Species

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ABSTRACT

The accuracy of tracer gas method depends vitally on the spatial uniformity of tracer/air mixing, which in many cases is unsatisfactory. However, information on this critical problem has been scarce, largely due to the practical difficulty in experimentally obtaining data. This paper presents a study of the effect of tracer species on tracer mixing, using a time dependent CFD method. It was found that for single-zone tracer decay tests, three tracer-gases, sulphur hexafluoride, nitrous oxide and carbon dioxide have virtually identical mixing patterns and thus there is no difference between them in terms of airflow measurement results. However, for multi-gas tests where there is interzonal tracer movement, the tracer-gases with different binary diffusivities exhibit significantly different mixing behaviour. In these situations, the choice of tracer will impact the accuracy of airflow measurement. These results support recent experimental findings concerning the significant effect of tracer species on airflow measurement.

NOTATION

A, B	Components A and B
C	Tracer concentration (%)
D	Molecular diffusivity (m^2/s)
k	Boltzmann's constant, 1.3805×10^{-25} (J/K)
M	Molecular weight (g/mol)
P	Pressure (Pa)
t	time (second)
T	Temperature (°C)

T_b	Normal boiling point (°C)
V	Velocity
V_b	Liquid molar volume at boiling point (m^3/mol)
ε	Characteristic energy parameter
Ω_D	Diffusion collision integral (dimensionless)
μ_p	Dipole moment (debyes)
σ	Collision diameter (m)

INTRODUCTION

Building energy consumption accounts for up to 50% of all energy use and building energy loss due to air infiltration and ventilation is increasingly significant as the building envelop becomes better thermally insulated. There is obviously a need for accurate measurement of the air exchange rate between buildings and the outside environment. Tracer-gas techniques[1], including the concentration-decay, constant-concentration, constant-injection, pulse-injection and the passive PFT technique are the most widely used methods for this type of measurements. They do not require the knowledge of the air inlet positions which are usually unknown and they are more accurate than other techniques at low flow velocities, which are usual in buildings. There have been significant advances in the development and wider application of these techniques[2-5]. Nevertheless, there is a potential problem related to all versions of tracer-gas techniques, i.e. the discrepancy between the less than perfect tracer mixing achieved in practical tests and the theoretical requirement of uniform tracer concentration within the test zone during the test. This requirement implies that the supply air entering the zone must instantly achieve uniform mixing with the air-tracer mixture in the zone. This is physically unsound and mixing enhancement is needed in most test situations to reasonably satisfy the above requirement. The most popular method for this purpose is the use of a mixing fan. However, it is not universally applicable.[1, 6] For example, in the measurement of natural convective air infiltration, the driving force of the flow is weak in relation to the fan jet, which may disturb the flow rate to be measured. Although some alleviation of this problem can be effected by properly positioning the fan so that its flow does not directly impinge on the inlet or outlet of the flow being measured, the fact that many flows being measured are with unknown inlet or outlet positions make this difficult to implement in practice.

As a result, the reliable use of mixing fans for tracer mixing enhancement is confined to a limited number of situations and tracer mixing is still, in general, an unsolved difficulty.

The accuracy of tracer-gas methods has been the subject of much research. However, until recently it has been predominantly along the line of analysing the effect of measurement errors on the flow rate results. Such work provided information on acceptable measurement errors and elucidated the most error tolerating algorithm for flow rate derivation. These studies, by accepting the algorithms, implicitly assume that the tracer concentration is uniform and errors arise solely from equipment or operators. They therefore did not address the crucial problem of mixing. More recently, the focus of research is starting to switch to the more fundamental study of factors affecting tracer mixing[6-9]. These are necessary in order to provide insight into the mixing mechanisms, based upon which, better mixing enhancement methods may be devised in the future. It has been shown that smaller building zones, lower air change rates and higher inlet airflow velocities have positive effects on tracer/air mixing and that there does not exist a universal critical value of air change rate below which satisfactory mixing is guaranteed. Nevertheless, many more questions regarding tracer mixing remain unanswered.

This paper presents a study of the effect of tracer species on mixing. It is usually assumed[1, 6] that tracer mixing is independent of the tracer species, which appears reasonable, given the always small proportions of tracer-gas in tracer-air mixtures. However, there is recent experimental evidence[8, 9] to contradict this assumption. In both studies, three different tracer-gases were used and these gave rise to different flow rate results for the same flow. This study examines this problem using an analytical/computational approach. Analytical results were used firstly to compare with the experimental findings with a view to verifying the latter and secondly to provide a theoretical explanation of the effect.

COMPUTATION PROCEDURES

The CFD code FLUENT was used to solve the three dimensional Navier-Stokes equations, i.e. the momentum equations, the continuity equation and the mass transfer (for the tracer-gas) equation. To simulate the transient tracer concentration variation, the time dependent versions of the above equations were used.

Computational Domain and Boundary Conditions

The building zone used in this study, or the computation domain, is of cubic shape and measures 3m×3m×3m (Fig. 1). The three-dimensional computational domain was selected in preference to the two-dimensional one, despite the penalty in CPU processing time. It is felt that in a two-dimensional domain, the air-stream from the inlet to the outlet will divide the domain into separate areas, isolating them from each other, resulting in mixing being artificially obstructed. The zone used in the computation is symmetrical about its central symmetrical plane. This fact was utilised to reduce the amount of computer storage and calculation required by including only one half of the zone in the computation domain. As a consequence, the central symmetrical plane becomes one of the boundaries and the symmetrical boundary conditions were imposed, which assumes that the velocity component perpendicular to the plain is zero and all scalar gradients are zero. The supply air inlet for the room, as shown in Fig. 1, has a square cross section (0.3m × 0.3m). The supply air stream enters the room perpendicular to the inlet at a uniform speed of 0.01 m/s. This resembles the magnitude of stack-driven natural convection between building zones and provides a bulk airflow rate of 1/3 ach. This velocity was selected to enable the computational assessment of tracer mixing in zones that are not saturated with fully developed turbulence. In situations where fully developed turbulence prevails, the mixing between air and different species of tracer will be identical. This is because molecular mixing which is species-dependent is several orders of magnitude weaker than turbulent mixing, the latter being dependent only on the flow properties. However it is rare to find fully developed turbulence in building air infiltration and ventilation. In the vast majority of cases, there is co-existence of turbulent areas and laminar areas or the flow is completely laminar. Thus there could be differences in terms of mixing between different species. The computational verification of the effects of tracer species is complicated by the fact that the currently available commercial CFD codes can not deal with mixed turbulent and laminar flows. Therefore, the above set-up was chosen to ensure that the flow throughout the zone is laminar and that the mixing is thus accurately simulated. As will be shown clearly in the section "Results and Discussion", this choice is satisfactory for the purpose of this study. The outlet for the exhaust air has the same shape and dimensions as the inlet and only half of both were included in the computation domain as is the case for the building zone.

Numerical Scheme

The Navier-Stokes equations were discretised into finite volume equations, based on three-dimensional Cartesian grids. Denser grids were used closer to the corners and jet boundaries and coarser ones away from these features to limit the total number of cells. The expansion rate of the distance between grid lines was restricted to below 1.2 to promote numerical stability. The computations were time dependent to deal with the tracer-concentration decay. Small time steps, around 1/10th of the characteristic time scale based on inlet air velocity, were used and the flow fields at 330 time-steps were computed. The equations resulting from the discretisation process were solved using the SIMPLE algorithm. The solution convergence speed was accelerated by means of a multi-grid technique in which four levels of grids with increasing coarseness were utilised. At the end of each time step, the normalised residuals for the equations were around 10^{-6}.

Binary Diffusivity

Three tracer-gases, sulphur hexafluoride, nitrous oxide and carbon dioxide, were used to examine the effect of tracer species on mixing. Diffusion of one gaseous species into another is governed by

$$J = -D \frac{dC}{dx},$$

for one-dimensional diffusion and the equivalent equation for three-dimensional diffusion is very similar. Here J is the diffusion flux across unit area normal to the x-direction, $\frac{dC}{dx}$ is the concentration gradient and D is binary diffusivity. There are limited experimental data for binary diffusivities because of the vast number of possible binary combinations of species and the excessive amount of measurement that would be required. Binary diffusivity data for the three tracer-gases used in this computation were calculated based on molecular kinetics[10] as outlined in the following. Accuracy of such calculation is around 5 percent. The binary diffusivity of a two gaseous species (A and B) system is given by:

$$D_{AB} = 1.858 \times 10^{-3} \, T^{3/2} \frac{(\frac{1}{M_A} + \frac{1}{M_B})^{1/2}}{P \sigma_{AB}^2 \Omega_D},$$

where

M = molecular weight, g/mol

D_{AB} = binary diffusivity, cm^2/s

T = temperature, K

P = pressure, atm

Ω_D is diffusion collision integral (dimensionless) which can be calculated using

$$\Omega_D = \frac{A}{T^{*B}} + \frac{C}{e^{DT^*}} + \frac{E}{e^{FT^*}} + \frac{G}{e^{HT^*}} + \frac{0.19\delta_{AB}^2}{T^*} \qquad (1)$$

where A = 1.06036, B = 0.15610, C = 0.19300, D = 0.47635, E = 1.03587, F = 1.52996, G = 1.76474, H = 3.89411;

$$\delta_{AB} = (\delta_A \delta_B)^{1/2}$$

$$T^* = \frac{kT}{\varepsilon_{AB}}$$

$$\frac{\varepsilon_{AB}}{k} = (\frac{\varepsilon_A}{k} \frac{\varepsilon_B}{k})^{1/2}$$

$$\frac{\varepsilon}{k} = 1.18(1 + 1.3\delta^2)T_b$$

$$\delta = \frac{1.94 \times 10^3 \mu_p^2}{V_b T_b}$$

μ_p^2 = dipole moment, debyes

V_b = liquid molar volume at boiling point, cm^3/g-mol

T_b = normal boiling point, K

k = Boltzmann's constant, 1.3805×10^{-25} J/K

T = temperature, K

σ_{AB} in equation (1) is defined by

$$\sigma_{AB} = (\sigma_A \sigma_B)^{1/2}$$

where σ is collision diameter with unit $\overset{\circ}{A}$ and is evaluated by

$$\sigma = (\frac{1.585 V_b}{1 + 1.3\delta^2})^{1/3}$$

The binary diffusivies for sulphur hexafluoride-air, nitrous oxide-air and carbon dioxide-air at the temperature of 288K were calculated as 8.8532×10^{-6}, 1.35566×10^{-5} and 1.50348×10^{-5} m^2/s.

Procedure

Single-tracer or multiple-tracer decay tests can be set up in a variety of ways and lead to a variety of tracer mixing patterns, which can be classified into two groups. Consider the simplest multiple-tracer decay tests: Two zones, A and B, are injected with tracers "a" and "b" respectively. During the test, fresh air from the outside environment and flow from zone B will mix with the tracer "a" and air mixture in zone A, diluting tracer "a". On the other hand flow from zone B also carries tracer "b" into zone A, increasing the "b" concentration there. These two distinct mixing situations occur in a similar way to zone B. Indeed, the two groups of mixing situation can be identified in all multiple tracer tests. The first group is typified by the case of fresh air entering a zone containing tracer-air mixture. Mixing in all single-tracer decay tests and mixing in parts of multiple tracer decay tests (e.g. mixing of tracer "a" in zone A in the above example) belong to this group, which is referred to as type "aA" mixing in the following. The second group is typified by tracer-air mixture entering a zone of fresh air. Mixing of tracer "b" in zone A in the above example belongs to this group, which is referred to as type "bA" mixing in the following. Obviously the study of tracer mixing in decay tests should be centred on the examination of the above two typical cases.

In light of the above discussion, two groups of a total of six cases of tracer mixing were examined. In the first group, which corresponds to the "aA" type described above, the zone described in the section *"Computation Domain"* was injected with one of the three tracers described in the section *"Binary Diffusivity"* which then is mixed with the air in the zone to achieve a uniform concentration of 0.1%. As the test starts, fresh air enters the zone and the variation of tracer concentration distribution with time is recorded; The computation is halted when the accumulative air change reaches 1/3 ac. The same procedure is repeated for the other two tracer-gases and the three set of results are then compared to determine the effect of tracer species. The second group, corresponding to the "bA" type mixing, of three cases was computed in a similar manner. The only difference is that the air in the zone at the start of the tests is fresh (free from tracers) while the supply air entering the zone had a uniform tracer concentration of 0.1%

RESULTS AND DISCUSSION

Figs. 2, 3 and 4 are results from the first group of three cases which correspond to the "aA" type mixing as described in the section *"Procedure"*. The supply air contains no tracer-gas and the zone has a uniform initial tracer concentration of 0.1%. Fig. 2 shows the sulphur hexafluoride concentration distribution, by means of concentration contours, across the central symmetrical plane. This is a "snap-shot" at the end of the test when 1/3 air change has been accumulated. The concentration is measured as the ratio of the mass of the tracer to the mass of air. The values for the contours, in the order from upper-right to lower-left are, in equal steps, 9.93×10^{-4}, 9.41×10^{-4}, 8.89×10^{-4}, 8.36×10^{-4}, 7.84×10^{-4}, 7.32×10^{-4}, 6.80×10^{-4}, 6.27×10^{-4}, 5.75×10^{-4}, 5.23×10^{-4}, 4.70×10^{-4}, 4.18×10^{-4}, 3.66×10^{-4}, 3.14×10^{-4}, 2.61×10^{-4}, 2.09×10^{-4}, 1.57×10^{-4}, 1.05×10^{-4}, 5.23×10^{-5}, respectively. Figs. 3 and 4 are interpreted in the same way except that they show the concentration distribution of nitrous oxide and carbon dioxide, respectively. The distribution patterns show remarkable similarity. At any particular point on the plane the differences in concentration between the three tracer-gases are smaller than 5%. In fact, the similarity is repeated across the complete zone. These results show that the mixing between air and each of the three tracer-gases is virtually identical and that all three tracer-gases would yield the similar results when used in flow rate measurement. Although the conclusion is derived from cases of laminar mixing, it can be generalised to include other flow conditions. As will be described in the following, there are three mechanisms of mixing: diffusion, convection, and turbulence. Diffusion is dependent on two factors, concentration gradient and molecular diffusivity; convective mixing is governed by velocity field and concentration gradient. Thus mixing is determined by four factors: concentration gradient, molecular diffusivity, velocity field and turbulence. Since the concentration distribution is identical at the start for all three tracers, the difference thereafter in concentration gradient between them must be due to the latter three factors. The determining factors are therefore reduced to three. In the above three cases, the flow fields are identical and turbulence is absent. Therefore, the close similarity between the mixing patterns implies that in Group 1 situations, the molecular diffusivity has little effect on mixing. This together with the fact that velocity field and turbulence are independent of tracer species, provided tracer concentration is small as is true for virtually all situations, dictates that in Group 1 situations the choice of tracer species will not affect flow measurement results for laminar or turbulent flows.

Very different results emerge from the second group of three cases which correspond to the "bA" type mixing" as described in the section *"Procedure"*. The supply air has a uniform tracer concentration of 0.1% and the zone contains no tracer-gas at the start of the test. Fig. 5 shows the tracer concentration histories for sulphur hexafluoride, nitrous oxide and carbon dioxide at a spatial point with co-ordinates in x, y and z axes of 0.5, 2.5 and 1.5, respectively. The definition of the co-ordinate system is shown in Fig. 6. Large differences in concentration, especially between sulphur hexafluoride and the other two tracers, is evident and maintained throughout the test duration. These differences are consistent with the fact that the binary diffusivities of nitrous oxide and carbon dioxide (10.9% difference between them) are much larger (70% based on CO_2) than that of sulphur hexafluoride. Obviously, the mixing performances of the three tracer-gases are significantly different, which is closely related to their widely differing molecular diffusion capacity.

The histories of concentration variation of the three tracer-gases at 19 other points were also examined. The position of the 20 sampling points are shown in Fig. 6. Because of the symmetrical nature of the zone, sampling points from only one half of the zone need be selected. These points are located on three planes vertical to the z-axis. One of the planes is the central symmetrical plane, the second is close to the wall (z=0.05m) and the other is in between the two with z=0.5m. To facilitate the appreciation of the spatial positions of the sampling points, those on the symmetrical plane and those on the plane close to the wall are marked with "north-east" pointing short lines and "south-west" pointing short lines, respectively. The twenty points are classified into three groups according to the gaps between the concentration curves for the three tracers. In the first group, the curves for the three tracers virtually coincide; Sampling points where the gap, in terms of relative difference in concentration, averages up to 10% are assigned to the second group and those with gaps over 10% form the third group. For example, the sampling point for Fig. 5 belongs to group three. Points of the three groups are assigned symbols of square, circle and triangle, respectively. As shown in Fig. 6, among the 20 sampling points, there are 3 in group one, 8 in group two and 9 in group three. These results indicate that tracer-air mixing for sulphur hexafluoride, nitrous oxide and carbon dioxide are significantly different for "bA" type situations.

It has been explained in the section *"Procedure"* that all single-zone tracer decay tests are associated with "aA" type tracer mixing and all multi-zone or multi-tracer decay tests involve "bA" as well as "aA" type mixing. It follows from the above results concerning the two mixing types that for single-zone tracer decay tests, there is no difference between the three tracer-gases, sulphur hexafluoride, nitrous oxide and carbon dioxide, in terms of results of flow rate measurements. However, for multi-tracer-gas tests, the choice of tracer will impact the result of airflow measurement. This conclusion supports recent experimental findings by Kohal and Riffat[9] who revealed the significant effect of tracer species on flow rate measurement results. They revealed that nitrous oxide tends to perform better than sulphur hexafluoride, which is in line with the fact that the binary diffusivity of the former is significantly higher and that tracer concentration distribution results from this study show greater uniformity for the former. The results from this study also show that carbon dioxide tends to have better mixing than nitrous oxide and sulphur hexafluoride, as it has the highest binary diffusivity of the three tracer-gases. However, significant and variable background concentration and relatively poor detectability of carbon dioxide discourage its use as a tracer-gas. The level of detectability of carbon dioxide is around 3 and 7 orders of magnitudes lower than those of nitrous oxide and sulphur hexafluoride[1], respectively. As a result, 100 litres of carbon dioxide may need to be injected into a modest sized (3m×3m×3m) zone. Heating, cooking and breathing contribute to oscillation in background carbon dioxide concentration, causing uncertainty and inaccuracy in flow rate measurements. As a result, carbon dioxide may not perform as well as the other two tracers despite of its high diffusivity. This is borne out by recent experimental results[9].

Fig. 7 shows the tracer concentration histories for sulphur hexafluoride, nitrous oxide and carbon dioxide at a point (Group One) with co-ordinates of 2.5, 0.5 and 0.5, respectively. The curves for the three tracer-gases virtually coincide with one another. Although this is a relatively rare occurrence, it does provides an insight into the mixing mechanism: it shows that molecular diffusivity is not the only determining factor, even in laminar mixing situations. The close resemblance of the curves for the three gases indicates the existence of an active mixing mechanism in addition to molecular diffusion: in this case, convective mixing. Transient tracer transport as in the cases being studied, is governed by

$$\frac{\partial C}{\partial t} = -V\frac{\partial C}{\partial x} - C\frac{\partial V}{\partial x} - D\frac{\partial^2 C}{\partial x^2},$$

for one-dimensional transport (the equation for three-dimensional transport is similar) where

C = tracer concentration

t = time

V = velocity

D = molecular diffusivity

x = one dimensional co-ordinate

Because there is negligible mass accumulation at any point in the flow field in tracer test situations, the second term on the right side of the equation is negligible. Thus the above equation reduces to

$$\frac{\partial C}{\partial t} = -V\frac{\partial C}{\partial x} - D\frac{\partial^2 C}{\partial x^2}$$

The two mechanisms are clearly shown in the equation: diffusive mixing by the second term on the right side and convective mixing by the first term on the right. It is clear that convective tracer mixing is directly proportional to velocity magnitude. However, high velocity magnitude is not sufficient; the direction of velocity must be in relatively the same direction as that of tracer concentration gradient for convective mixing to be effective. In the situation where the two directions are perpendicular to each other, convective mixing will not occur at all. In fact this is what happened to the case shown in Fig. 5 where the downward flow direction is normal to that of the local tracer-concentration gradient, which can be appreciated by examining Figs. 2-4. Consequently molecular diffusion becomes dominant and the differences in binary diffusivity between the tracers were manifested in the mixing result. On the other hand, places with relatively large velocity in the appropriate direction will have considerable convective mixing, of the same order of magnitude as the diffusive mixing in the case depicted in Fig. 7. The above discussion also serve to illustrate the role played by three of the four contributing factors of tracer mixing, which are tracer-concentration gradient, velocity field, tracer molecular diffusivity and, not discussed so far, turbulence.

Turbulent mixing is intensified convection. Its effect is reflected by the parameter of turbulent diffusivity, which is a function of flow properties and usually several orders of magnitude greater

than molecular diffusivity. These two factors ensure that molecular diffusivity, and thus the choice of tracer, will not affect the concentration distribution or mixing of tracer. However, airflow in building zones are not usually fully turbulent. Buildings without mechanical ventilation are dominated by natural convective ventilation where the Reynolds numbers are in the order of magnitude of 10^3, as in the above simulated cases and the flow is laminar. Even in mechanically ventilated buildings, depending on the air supply rate and the location in a building, the flow may not be turbulent all the time or everywhere. This investigation by selecting a low Reynolds number situation and avoiding using the turbulence model, has sought to ascertain whether the choice of tracer-gas can affect the result obtained using tracer-gas technique, in the vast range of non-turbulent situations. The answer is positive.

CONCLUSIONS

The effect on mixing of tracer species has been examined using an analytical/computational approach. The distribution and history of tracer concentration during airflow measurements were obtained using Computational Fluid Dynamics (CFD). It was found that for single-zone tracer decay tests, three tracer-gases, sulphur hexafluoride, nitrous oxide and carbon dioxide have virtually identical mixing patterns and thus there is no difference between them in terms of flow rate measurement results. However, for multi-tracer-gas tests where there is interzonal tracer movement, the three tracer-gases with different binary diffusivities exhibit significantly different mixing behaviour. In these situations, the choice of tracer will impact the accuracy of airflow measurement. These conclusions support recent experimental findings concerning the significant effect of tracer species on flow rate measurement results.

The sequence of the tracers, in the order of descending mixing power, is carbon dioxide, nitrous oxide and sulphur hexafluoride, which is a result of their different diffusivities, also in the same order. However, mixing power should be considered together with other practical factors when making a choice of tracer-gas. Use of carbon dioxide should be avoided despite of its high diffusivity because of its significant and variable background concentration and relatively poor detectability.

It should be pointed out that in the relatively rare situation where airflow throughout the building zones being tested is turbulent, the mixing performance and measurement results will be identical for the tracer-gases.

It is clear from the analysis process that although the study was based on three gases, the conclusions given above in the first paragraph apply to all tracer-gases as the effect of tracers is manifested only via their diffusivities.

The investigation concentrated on tracer mixing involved in the decay technique. For other tracer-gas techniques, the "aA" type mixing will be significantly different. Multiple injection points, which does not apply in the decay technique, are routinely used for the constant-injection and constant-concentration techniques and may influence mixing. More investigation is necessary to determine whether the above conclusion regarding the single tracer technique applies to the latter two techniques. Close examination of "bA" type mixing shows that is unaffected by the latter two types of tracer-gas techniques. It follows that measurement results of multi-zone constant-concentration and constant-injection techniques are affected by the choice of tracer.

The effect on tracer mixing of many factors remains unclear. For example, further research effort is necessary to ascertain the effect of building zone geometry and the effect of mixing fans on mixing enhancement.

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Fig. 1   Schematic of building zone used in computation.

Fig. 2   Concentration distribution of sulphur hexafluoride.

Fig. 3   Concentration distribution of nitrous oxide.

Fig. 4   Concentration distribution of carbon dioxide.

Fig. 5. Concentration histories for three tracer-gases at point (0.5, 2.5, 1.5).

Fig. 6   Schematic of the locations of the twenty sample-points.

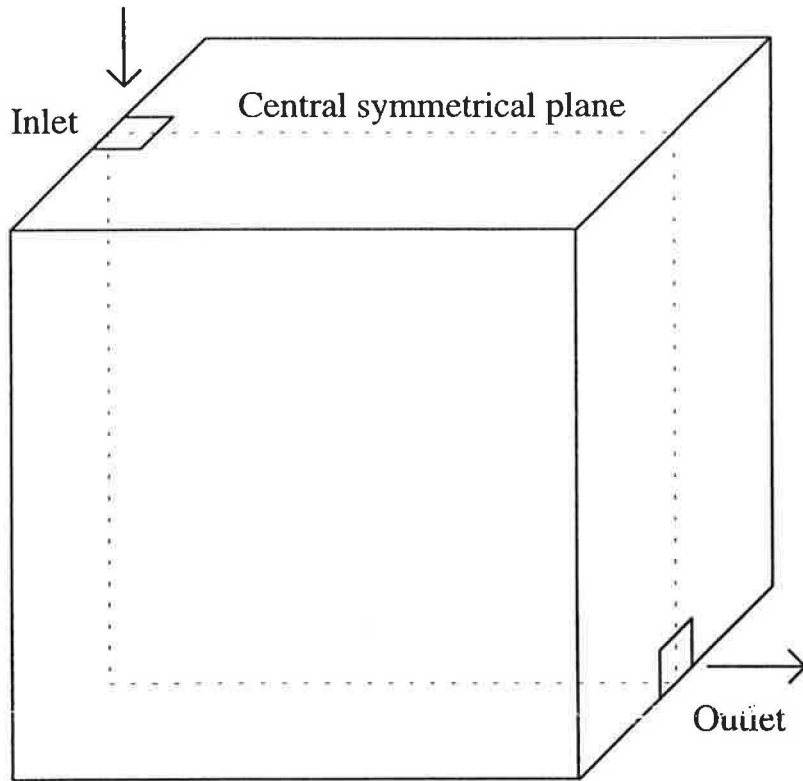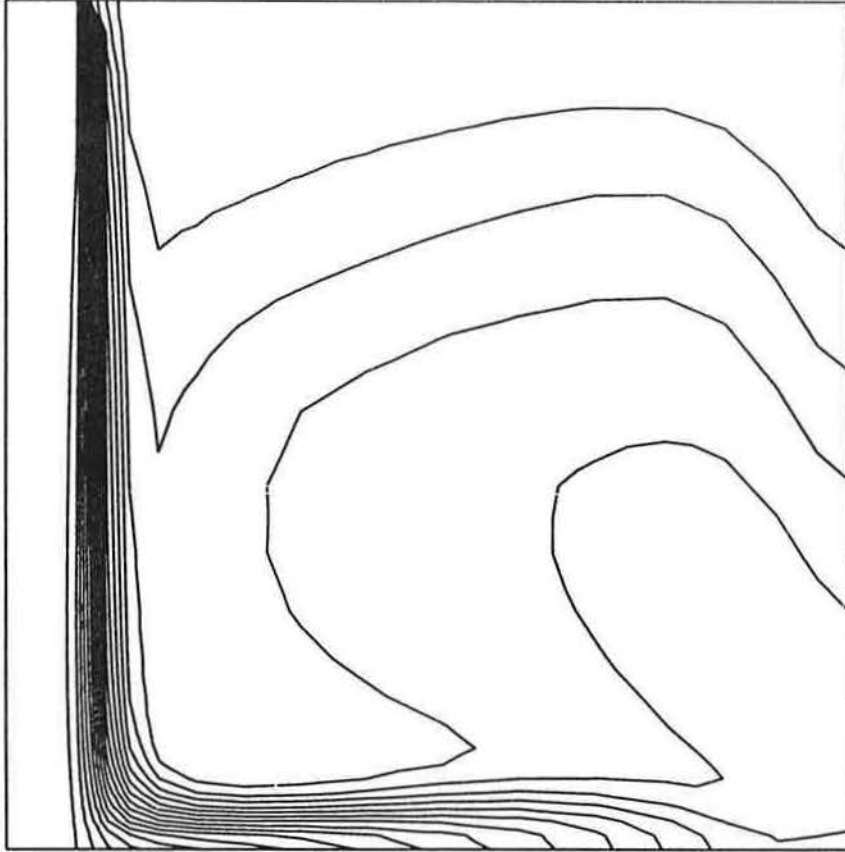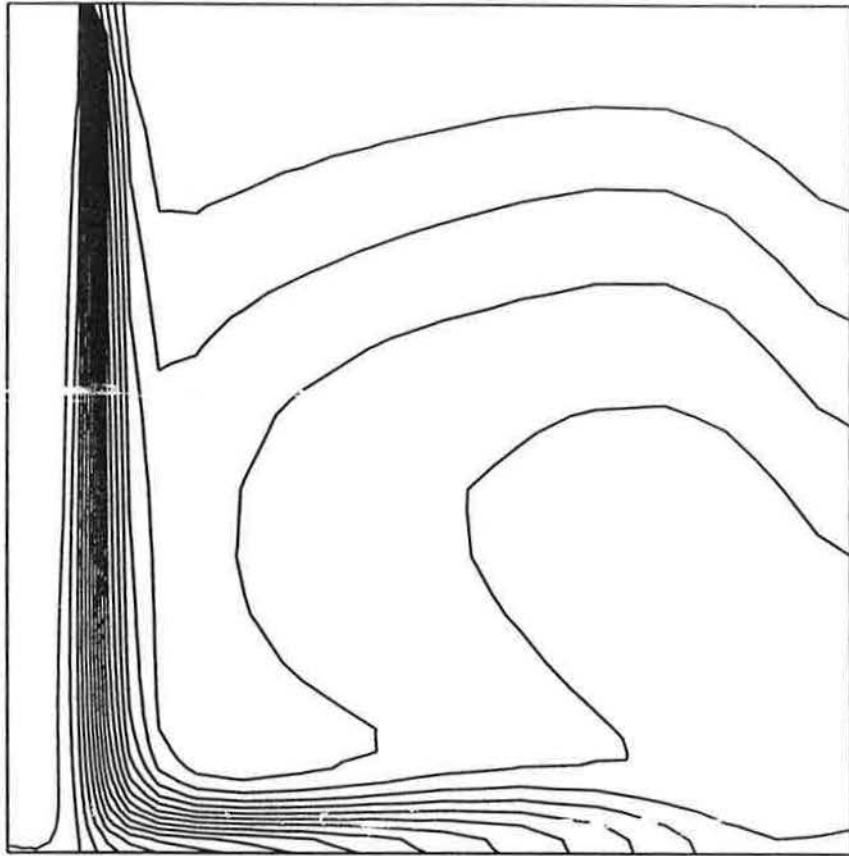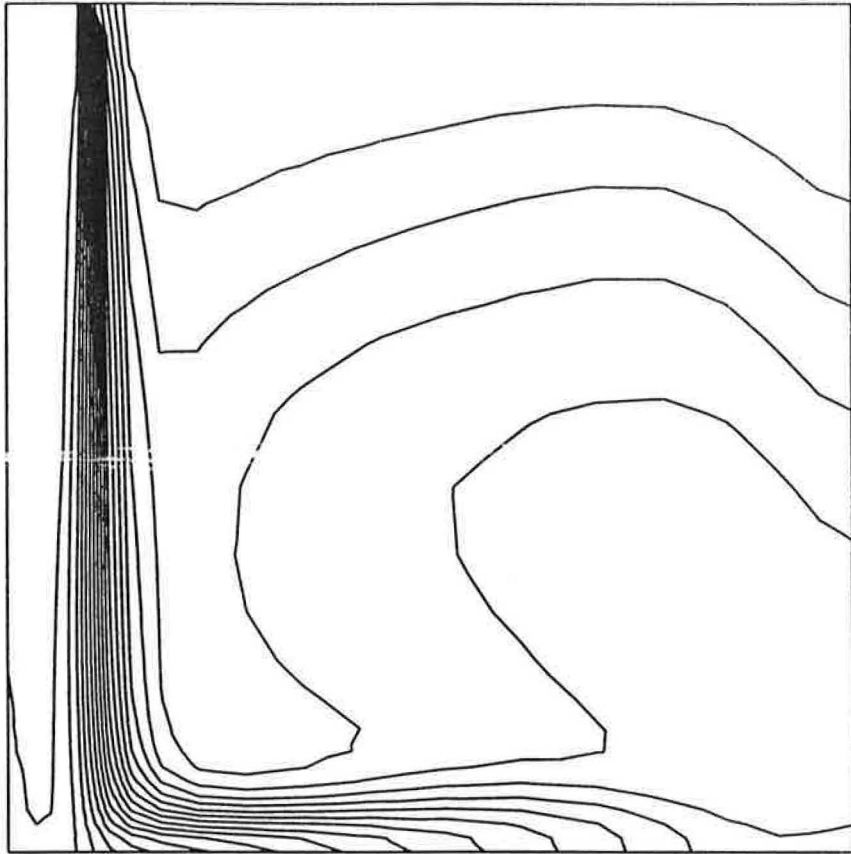Fig. 7. Concentration histories for three tracer-gases at point (2.5, 0.5, 0.5).

Inlet

Central symmetrical plane

Outlet

Fig. 1

15

Fig 2

Fig 3

Fig. 4

Fig. 5

Central symmetrical plane

Fig. 6

Fig. 7