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Consideration of Turbulent Deposition in Aerosol Behaviour Modelling with the CONTAIN Code and Comparison of the Computations to Sodium Release Experiments

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Abstract

Consideration of Turbulent Deposition in Aerosol Behaviour Modelling with the CONTAIN Code and Comparison of the Computations to Sodium Release Experiments

CONTAIN is a computer code to analyze physical, chemical and radiological processes inside the reactor containment in the sequence of a severe reactor accident. Modelling of the aerosol behaviour is included. We have improved the code by implementing a subroutine for turbulent deposition of aerosols. In contrast to previous calculations in which this effect was neglected, the computed results are in good agreement with sodium release experiments. If a typical friction velocity of 1 m/s is chosen, the computed aerosol mass median diameters and aerosol mass concentrations agree with the experimental results within a factor of 1.5 or 2, respectively. We have also found a good agreement between the CONTAIN calculations and results from other aerosol codes.

Zusammenfassung

Berücksichtigung der turbulenten Deposition bei der Modellierung des Aerosolverhaltens mit dem CONTAIN-Code und Vergleich der Rechnungen mit Natriumfreisetzungsexperimenten

CONTAIN ist ein Rechencodesystem zur Analyse physikalischer, chemischer und radiologischer Prozesse im Reaktorcontainment beim Ablauf eines schweren Störfalls. Die Modellierung des Aerosolverhaltens ist eingeschlossen. Wir haben den Code verbessert durch Einbau eines Unterprogramms, das die turbulente Deposition des Aerosols beschreibt. Im Gegensatz zu früheren Rechnungen, bei denen dieser Effekt vernachlässigt war, stimmen die Rechenergebnisse jetzt gut mit Natrium-Freisetzungsexperimenten überein. Wenn für die Reibungsgeschwindigkeit der typische Wert von 1 m/s angenommen wird, stimmen die berechneten mittleren Aerosol-Massendurchmesser und die Massenkonzentrationen mit den experimentellen Werten innerhalb eines Faktos 1,5 bzw. 2 überein. Weiterhin haben wir auch eine gute Übereinstimmung zwischen den CONTAIN-Rechnungen und den Ergebnissen von anderen Aerosolcodes festgestellt.

Contents

1. Introduction

The computer code CONTAIN has been developed at Sandia National Laboratories (Bergeron et al. 1985). It is an integrated tool for predicting the physical, chemical and radiological conditions inside the reactor containment building following the release of radioactive material from the primary system after a severe nuclear reactor accident. CONTAIN may be applied to light water reactors (LWR's) and to liquid-metal-cooled fast breeder reactors (LMFBR's). The aerosol part in CONTAIN is based on the MAEROS code (Gelbard and Seinfeld 1980). Using the version 04, we have performed CONTAIN calculations for sodium fire aerosols as well as for metallic sodium droplets in closed vessels and compared the computed aerosol mass concentrations and aerodynamic mass median diameters to the results from sodium release experiments. On this occasion, we found that some improvements for a realistic prediction of aerosol behaviour were necessary. In case of sodium fire, sodium evaporation and sodium spray fire experiments, a large gradient of temperature exists between the pool (temperatures between 400 and 900°C) and the atmosphere of the containment (temperature between 100 and 300°C) which leads to a significant effect of turbulent deposition. This has not yet been considered in CONTAIN. So, the computed aerosol mass concentrations exceeded the experimental values up to an order of magnitude (compare section 3). Therefore, the module of the PARDISEKO code to calculate turbulent deposition (Bunz 1984) was implemented into the CONTAIN program system. This module is based on theory and measurements of Sehmel (1973). The results of the improved model were again compared to sodium release experiments. This is subject of the present report. Our work was done in cooperation with the KfK Institute for Neutron and Reactor Physics with the aim of creating a KfK version of CONTAIN for the application to LMFBR safety analysis.

2. Computations without turbulence

The MAEROS code is based on the mass bilance equation for a multicomponent aerosol system in a closed vessel (Gelbard and Seinfeld 1980). The generalisation of this equation, which is solved in MAEROS, is

$$\frac{\partial}{\partial t} q_k(m,t) = \int_o^m d\mu\ \phi(\mu, m-\mu)\, q_k(\mu,t)\, C(m-\mu,t)$$

$$- q_k(m,t) \int_o^\infty d\mu\ \phi(\mu,m)\, C(\mu,t)$$

$$- R(m)\, q_k(m,t) + S_k(m) \qquad (1)$$

where q_k is the total airborne mass of component k per unit volume of particles with mass in the interval [m, m + dm], $\phi(\mu, m)$ is the coagulation rate between particles with mass μ and m (coagulation efficiency), C(m, t) is the number of particles per unit volume in the interval [m, m + dm], R (m) is the removal rate for particles with mass m and S_k (m) is the aerosol source term.

The first term in equation (1) characterizes the increase of particles in the mass interval [m, m + dm] due to coagulation, the second gives the loss caused by coagulation and the third describes the loss caused by other processes, e.g. by deposition.

The MAEROS code considers coagulation due to molecular diffusion, turbulence and gravitational settling and deposition caused by sedimentation and molecular diffusion. It is also possible to consider condensation and evaporation effects. However, this is not of significance in our case. The NALA sodium evaporation experiments which are discussed in this paper were performed at temperatures of 130°C at which the sodium vapour pressure is low and effects from the sodium vapour release can be neglected. In case of the FAUNA sodium fire experiments and the SOFICOV sodium spray fire experiments (section 3) we have sodium oxide aerosols. In all computations, a particle size interval between 0.1 and 100 µm is considered. The number of particle size classes was 40 with the exception of section 4. Herewith sufficiently accurate calculations were obtained (comp. section 4).

Fig. 1 shows sodium oxide aerosol mass concentrations computed with CONTAIN (version 04) in comparison to benchmark calculations for a large containment with size of a breeder reactor. The results for the different aerosol codes (ABC, AEROSIM, AEROSOLS, PARDISEKO) are taken from a study of the European community (Commission of the European Communities, 1984). The input data were:

Coagulation shape factor	= 1.5
Source duration	= 10 h
Geometrical standard deviation	= 2.0
Emission rate	= 2 t/h
Dynamic shape factor	= 1.5
Mass median radius	= 0.5 µm
Containment vessel volume	= 180 000 m^3
Gas temperature	= 100°C

Particle density	=	2.8 g/cm^3
Gas pressure	=	1 bar
Wall area	=	20000 m^2
Floor area	=	2800 m^2
Thermal conductivity ratio	=	$4 \cdot 10^{-2}$
Brownian boundary layer thickness	=	0.1 mm
Turbulent energy density dissipation rate	=	0 cm^2/ s^3

The influence of turbulence is not considered here, since data are not available.

Figure 1 shows that a good agreement exists between the CONTAIN code and the other aerosol codes, especially PARDISEKO (Bunz 1984). In the last case, the aerosol mass concentrations are not different by more than a factor of 1.6. A similar result is obtained for the aerosol mass median radii (fig. 2).

However, in case of sodium release experiments the agreement is worse. Because of the large gradient of temperatur, we have a significant effect of turbulence in the vessel. Omitting turbulent coagulation and turbulent deposition leads to an overestimation of the aerosol mass concentrations. In this connection, fig. 3 shows that the aerosol mass concentrations computed with CONTAIN exceed the experimental values from the NALA sodium evaporation experiment T7 (volume of the test vessel 2.2 m^3) (Sauter and Schütz 1983) up to an order of magnitude. A similar result is obtained for the FAUNA (fig. 4) and SOFICOV experiments, in which deviations up to a factor of 5 are found.

3. Computations with turbulence

Turbulent coagulation is considered in the CONTAIN code, but not turbulent deposition. The turbulent deposition rate depends on the friction velocity. It is calculated in the CONTAIN code from the turbulent energy dissipation rate as follows:

$$u_* = \sqrt[3]{E \cdot z} \qquad (2)$$

where z is the height of the containment (see Landau and Lifshitz, 1959, Pasquill 1962). E is a parameter to determine the turbulent coagulation rate (compare Commission of the European Communities 1984). It is entered as input into CONTAIN. We have chosen the turbulent energy dissipation rate in such a way that the friction velocity is 1 m/s. Referring to the results of Ialuria (1980) that the friction velocity is 20 to 50% of the convective stream velocity under natural convection, this corresponds to a stream velocity of 3.5 m/s which is typical for sodium release

experiments (Bunz and Sauter 1984). We have implemented the part of the PARDISEKO code (Bunz 1984) into CONTAIN which describes turbulent deposition. Now, the equations for the deposition coefficients (s^{-1}) concerning the wall, the floor and the ceiling of the containment are:

$$a_W = (v_{dif} + v_{thrml} + v_{VT}) \cdot \frac{A_W}{V} \tag{3}$$

$$a_F = (v_{dif} + v_{thrml} + v_{sed} \cdot C_T) \cdot \frac{A_F}{V} \tag{4}$$

$$a_C = (v_{dif} - v_{sed} \cdot C_T + v_{thrml} + v_{CT}) \cdot \frac{A_C}{V} \tag{5}$$

where v_{dif} is the diffusional deposition velocity, V_{thrml} the thermophoretical deposition velocity, A_W the area of the walls, A_F the area of the floor, A_C the area of the ceiling and V the volume of the chamber. The turbulent transport to the wall is described by

$$v_{VT} = u_* / Int \tag{6}$$

The diffusional resistance integral Int (see also equations (7) and (9)) has been calculated according to Sehmel (1973). It depends on the angle of the normal surface to the direction of gravity.

The turbulent stream to the ceiling is considered by

$$v_{CT} = \frac{v_{sed}}{exp(v_{sed} \cdot Int / u_*) - 1} \tag{7}$$

Since the sedimentation to the floor increases by the turbulent stream, the sedimentation velocity is replaced by

$$v_{ST} = v_{sed} \cdot C_T \tag{8}$$

with

$$C_T = (1 - exp(-v_{sed} \cdot Int / u_*^{-1})) \tag{9}$$

In the CONTAIN code, the particle mass and also the particle size changes in each interval during the integration of the deposition coefficients. To avoid a too long computation time, we have determined for each particle size class the median

values of the turbulent deposition velocities and correction factors and used these values for the integration.

To calculate the coagulation rate due to gravitational sedimentation, it is necessary to know the collision efficiency between particles of different sizes. In the version 04 of the CONTAIN code, the Fuchs formula (Fuchs 1951, 1964)

$$\varepsilon = \frac{3}{2} \cdot \left(\frac{r_2}{r_1 + r_2}\right)^2 \qquad (10)$$

with ε collision efficiency

r_1, r_2 radii of the particles ($r_1 > r_2$)

is used, which neglects the gravitational motion of the collected particle. We have replaced this formula by the more realistic formula of Pruppacher and Klett (Pruppacher and Klett, 1979) (see calculations in the fig. 3 - 5, 8 - 9)

$$\varepsilon = \frac{1}{2} \left(\frac{r_2}{r_1 + r_2}\right)^2 \qquad (11)$$

It considers both the velocity of the coagulating particle and the velocity of the collector.

In the NALA II experiments performed in a vessel with a volume of 2.2 m^3 and a height of 2.3 m, sodium aerosols were produced by evaporation from a hot liquid sodium pool at temperatures between 400 and 750°C. The vessel atmosphere was argon at 130°C. Aerosol mass concentrations were determined by impingers.

Figure 3 shows that the CONTAIN calculations are in good agreement with the results from the NALA experiment T7 after introducing the turbulent deposition. After a time of 1 h, the computed aerosol mass concentrations are not different from the experimental values by more than a factor of 2. The calculations (fig. 3) were performed under the assumption of a constant source rate over the heating time of 2.5 hours.

As shown in fig. 5, a good agreement is also found between measured and calculated aerodynamic mass median diameters. However, if the effect of turbulence is neglected, the computed diameters exceed the experimental values for NALA T3, T4 and T5 by about a factor of 2. This shows that especially the larger particles are deposited by turbulence. Omitting this effect leads to an increase of the aerodynamic mass median diameter.

Furthermore, our computations with turbulence show that the computed deposition rates agree well with the experimental results, too. From the NALA ex-

periment T7, it has been found that 89.9 g of the released sodium aerosols were deposited on the floor of the vessel and 16.1 g on the walls. Our computations yield 101.9 g for the floor and 6.5 g for the walls. Deposition on the ceiling is negligible in both cases.

Fig. 4 shows the aerosol mass concentrations for the FAUNA 2 sodium fire experiment performed in a test vessel with a volume of 220 m^3 (Cherdron and Jordan 1983). The burning time was 2.5 hours. The pool was heated up to temperatures between 400 and 950°C. The gas tempratures were approximately 200 °C. An automatically working sodium aerosol mass monitor was used which determined the aerosol mass concentrations continuously by a wet chemical method. For control purposes, measurements with impingers, filters and a mass balance were performed, too. Sodium oxide particles were produced which are nearly spherical. The dynamic shape factor was experimentally determined to 1.1. For our computations, we have varied the coagulation shape factor from 1 to 4. As shown in figure 4, the best adjustment of the aerosol mass concentrations is obtained if a dynamic shape factor of 1.1 and a coagulation shape factor of 2 is chosen. Then, the deviation between measured and calculated values is generally below a factor 2.

The PARDISEKO calculations predict the experimental data for a 2 m^2 pool experiment well with a dynamic shape factor of 1.1 and a coagulation shape factor of 4 (fig. 6). However, there are differences in the definitions. In PARDISEKO the effect of non-spherical shape on the particle's dynamic is considered by

$$X_{Par} = \frac{r_e^2}{r_{St}^2} \tag{12}$$

where r_e is the mass equivalent radius and r_{St} is the Stokes radius (Jordan et al. 1984). The coagulation shape factor is

$$\gamma_{Par} = \frac{r_c}{r_e} \tag{13}$$

with r_c collision radius (mean thrust radius).

The corresponding definitions used in CONTAIN (MAEROS) are (Gieseke et al. 1978)

$$X_{Mae} = \frac{R^2}{r_s^2} \tag{14}$$

$$\gamma_{Mae} = \frac{r_c}{R} \tag{15}$$

R is the radius of the agglomerate under the assumption that the density of the particle is changed from ρ to ρ' after agglomeration. r_s denotes the radius of the agglomerate having exactly a spherical shape at density ρ.The dynamic shape factor given by equation (14) can be interpreted as the ratio of the resistance of a given particle to that of a spherical particle having the same volume.

Furthermore, the agglomeration rate due to gravity is proportional to χ in PARDISEKO and to $\chi 2$ in CONTAIN. Turbulent coagulation which is influenced by the shape factors, too, is not considered in these PARDISEKO calculations (Bunz et al. 1988).

As a whole, it may be stated that both aerosol codes PARDISEKO and CONTAIN predict the values of sodium evaporation and sodium fire experiments well, if a friction velocity of about 100 cm/s is used, and agree well to each other (comp. fig. 4 to 7).

Concerning the aerodynamic mass median diameters, we have found that the values computed with CONTAIN are between 3 and 4 µm during the fire time and at 1.6 µm after a time of 20 hours. This is larger than the experimental results by about a factor of 1.5.

Finally, the aerosol mass concentrations computed with CONTAIN are compared to the large-scale SOFICOV (HEDL) sodium spray fire experiment SA 1 carried out by the Westinghouse Hanford Company (USA) in a vessel with a volume of 852 m^3 (Jeppson 1986). In this experiment, 650 kg of sodium at 541 °C were supplied for the spray fire. The fall height was 13.3 m to provide significant burning of the sodium during the fall. The gas temperatures were between 100 and 350 °C. Like in case of the FAUNA sodium fire experiments, sodium oxide particles were produced. The aerosol mass concentration was measured as a function of time by periodically passing a well-known quantity of gas through small filters located directly in the containment atmosphere and subsequent analyzing the material collected on the filter for sodium and total mass.

The diameter of the sodium spray fire droplets was determined by fitting it to the measured burning ratio. As expected, the best approximation of the experimental values is found, if a dynamic shape factor of 1.1 and a coagulation shape factor of about 2 is chosen. Figure 8 shows that for a time larger than 1000 s the aerosol mass concentrations computed with CONTAIN are even nearly identical with the experimental results, if the coagulation shape factor is 1.5. However, in this case the calculated values are too high for the beginning of the spray fire. The aerosol mass

concentrations computed with a coagulation shape factor of 2 are not different from the experimental curve by more than a factor of 2 in any case.

Furthermore, our computations show that the total sodium mass injected into the cell is deposited on the wall, the floor and the ceiling of the containment after a time of 2 hours (spray duration 2400 s).

However, the experimentalists detected only half of the masses. The reason may be that they investigated only a part of the surface area by water washing. The total deposited mass was determined by extrapolation which may lead to inaccuracies.

4. Variation of the number of particle size classes

The computing time of an aerosol computer code depends strongly on the number of particle size classes. The reduction of this number reduces the computing time considerably, but with a risk of numerical truncation leading to unrealistic results.

The smaller the number of particle size classes, the quicker decreases the aerosol mass concentration. According to Mitsutsuka et al. (1982), about 80 classes are needed to obtain reasonably accurate results for safety analysis. However, the MAEROS code is relatively unsensitive to the class number. Parameter studies according to Leigh and Helton (1984) show that a decrease of the aerosol size classes from 15 to 10 did not significantly alter the distribution functions for the integrated concentration, and only use of 5 sections altered the results by less than 10 per cent (time 10000 s). We have investigated this aspect, too, using a number of 10, 20, 40 and 94 size classes for our NALA computations (94 size classes is the maximum, at larger numbers the time steps for the Runge-Kutta integration routine become too small). Fig. 9 shows that the curves with 20, 40 and 94 classes are nearly identical (deviations less than 5 per cent). At the end of the computation where the difference is the largest, the concentrations computed with a number of 10 classes differ from the values with 20 classes by about 40 per cent. Thus, for safety reasons, we recommend to use a number of 40 classes for long time computations as we have done in sections 2 and 3. This leads to reasonable results, and the computation time is considerably shorter than for 94 particle size intervals (about a factor of 4 in case of the NALA experiment).

5. Conclusions

The aerosol part in the CONTAIN code basing on the MAEROS code was improved by implementing a subroutine which calculates the turbulent deposition according to theory and measurements of Sehmel (Subroutine Turbu). So it is possible to predict aerosol behaviour exactly even in case of a convective stream caused by a large gradient of temperature. The turbulent deposition rate depends on the friction velocity. The user must calculate or estimate the convective stream velocity from the gradient of temperature. Then, the friction velocity can be calculated under the assumption, that it is 20 to 50 per cent of the convection velocity (Ialuria, 1980). The friction velocity is determined in the computer code from the turbulent energy dissipation rate given as input data. We have compared the results from the new program version to sodium release experiments and found a good agreement between computations and experiments in all cases. The computed aerosol mass concentrations are not different from the results of sodium release experiments by more than a factor of 1.5 or 2, respectively. In the course of the FAUNA sodium fire and SOFICOV spray fire experiments, sodium oxide particles were generated which are nearly spherical. Our computations yield for these experiments the best approximation of the aerosol mass concentrations, if a coagulation shape factor of 2 and a dynamic shape factor of 1.1 is chosen. This is realistic in any case. A good agreement is also found between the CONTAIN calculations and the results of other aerosol codes, e.g. the PARDISEKO code (Bunz 1984). This is true even if turbulence is neglected. Thus, we can conclude that the CONTAIN code in the present version leads to a realistic simulation of aerosol behaviour and is a valuable tool for safety analysis.

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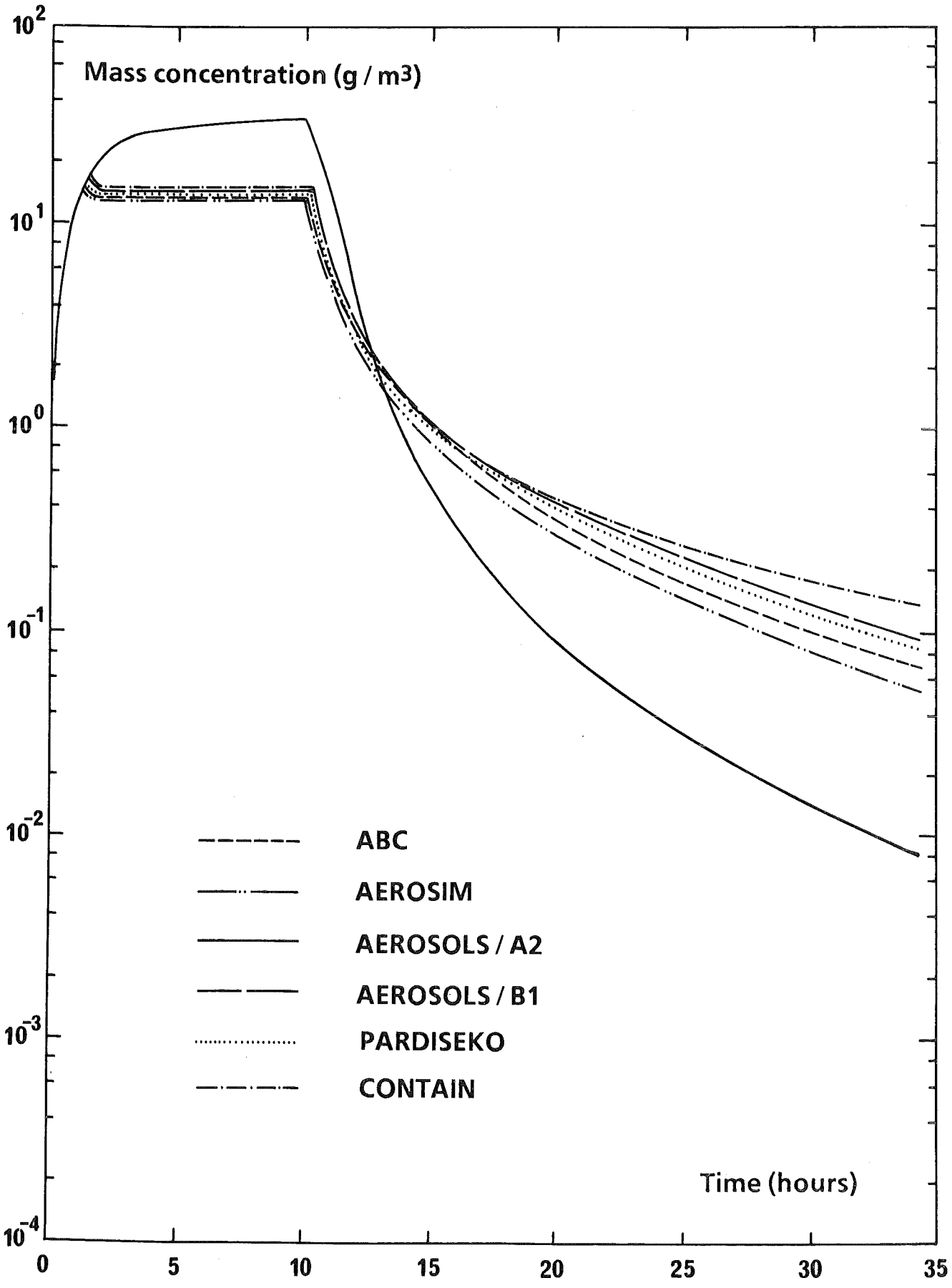

FIG. 1: SUSPENDED MASS CONCENTRATION FROM CALCULATIONS WITH DIFFERENT AEROSOL CODES

Reference Case (Fuchs $s = \gamma^2$)

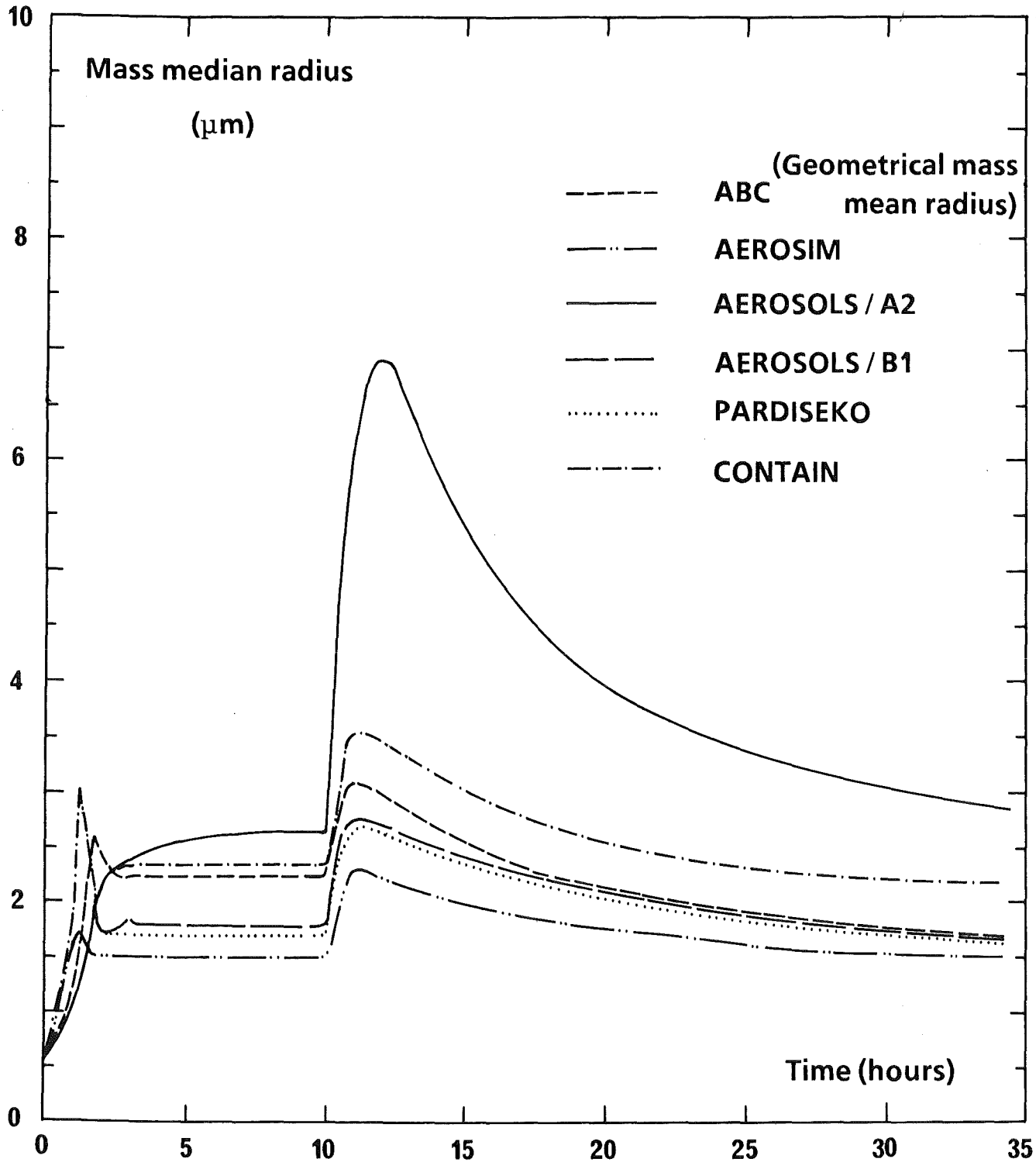

FIG. 2 : MASS MEDIAN RADIUS FROM CALCULATIONS WITH DIFFERENT AEROSOL CODES

Reference Case (Fuchs s = γ^2)

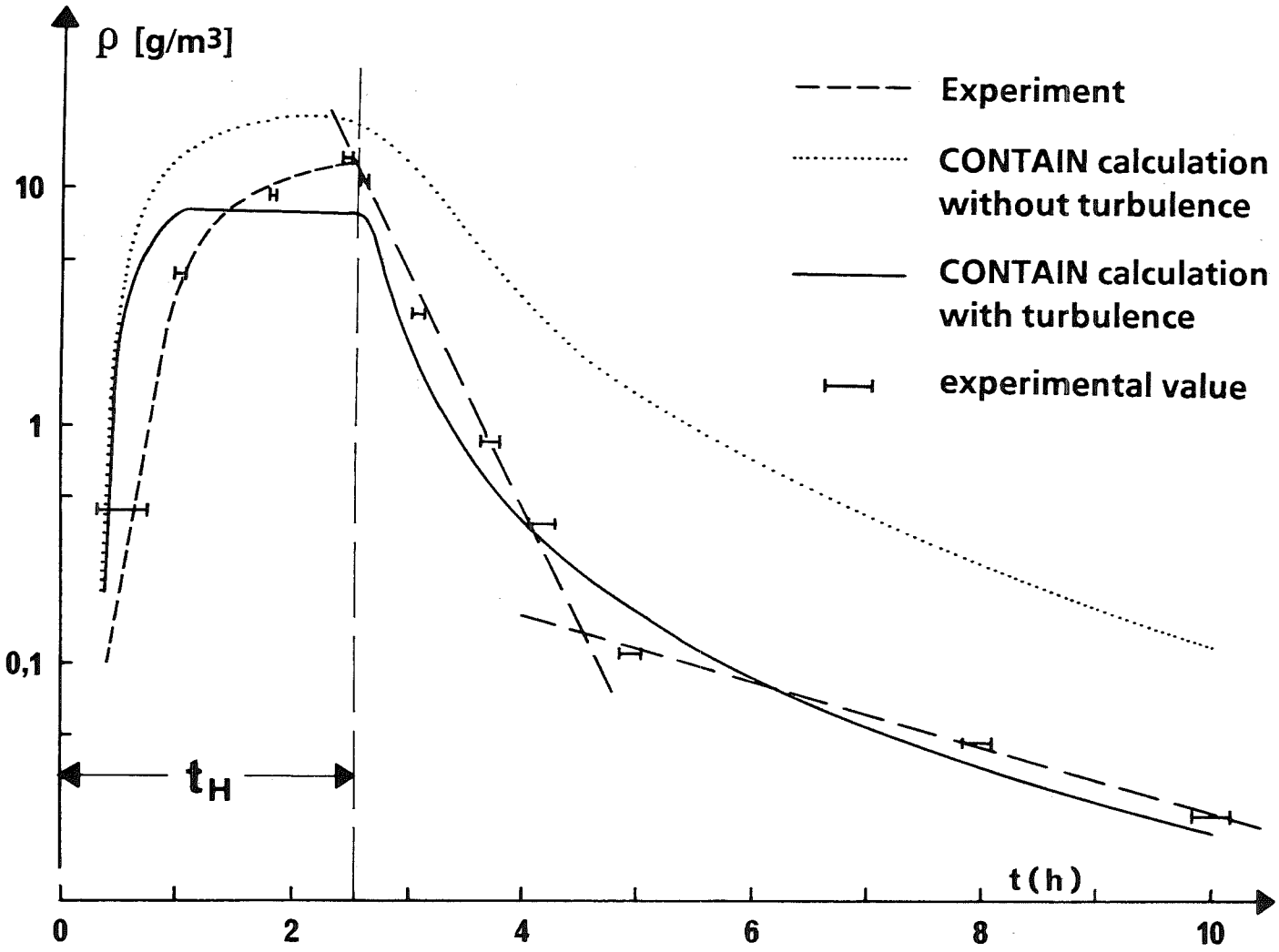

FIG. 3: SODIUM AEROSOL MASS CONCENTRATION FOR THE NALA EXPERIMENT T 7 IN COMPARISON TO CONTAIN CALCULATIONS (VOLUME OF THE TEST VESSEL = 2.2 m3)

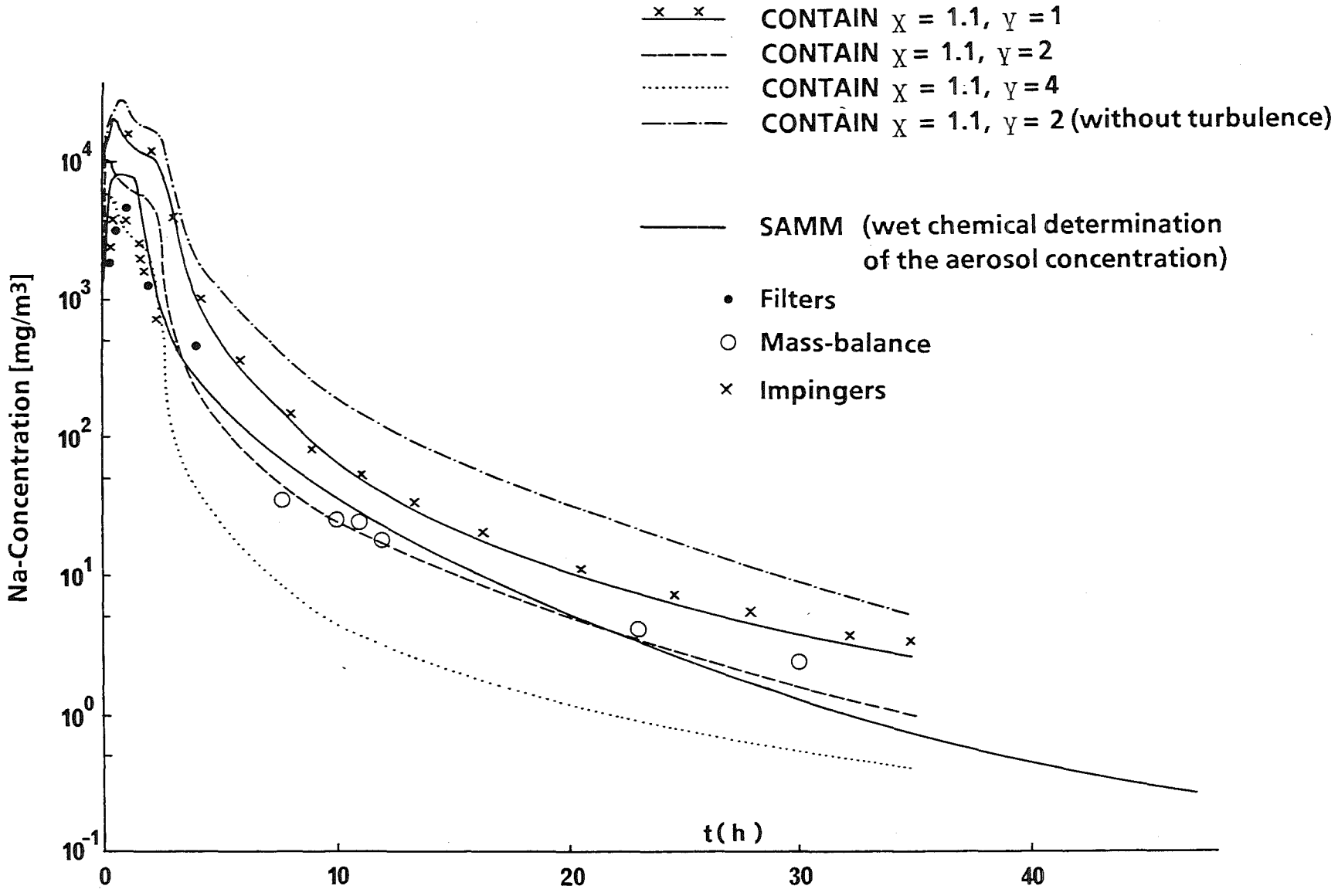

FIG. 4: AEROSOL MASS CONCENTRATION FOR THE FAUNA 2 SODIUM FIRE EXPERIMENTS (DURATION OF FIRE 2.5 h)

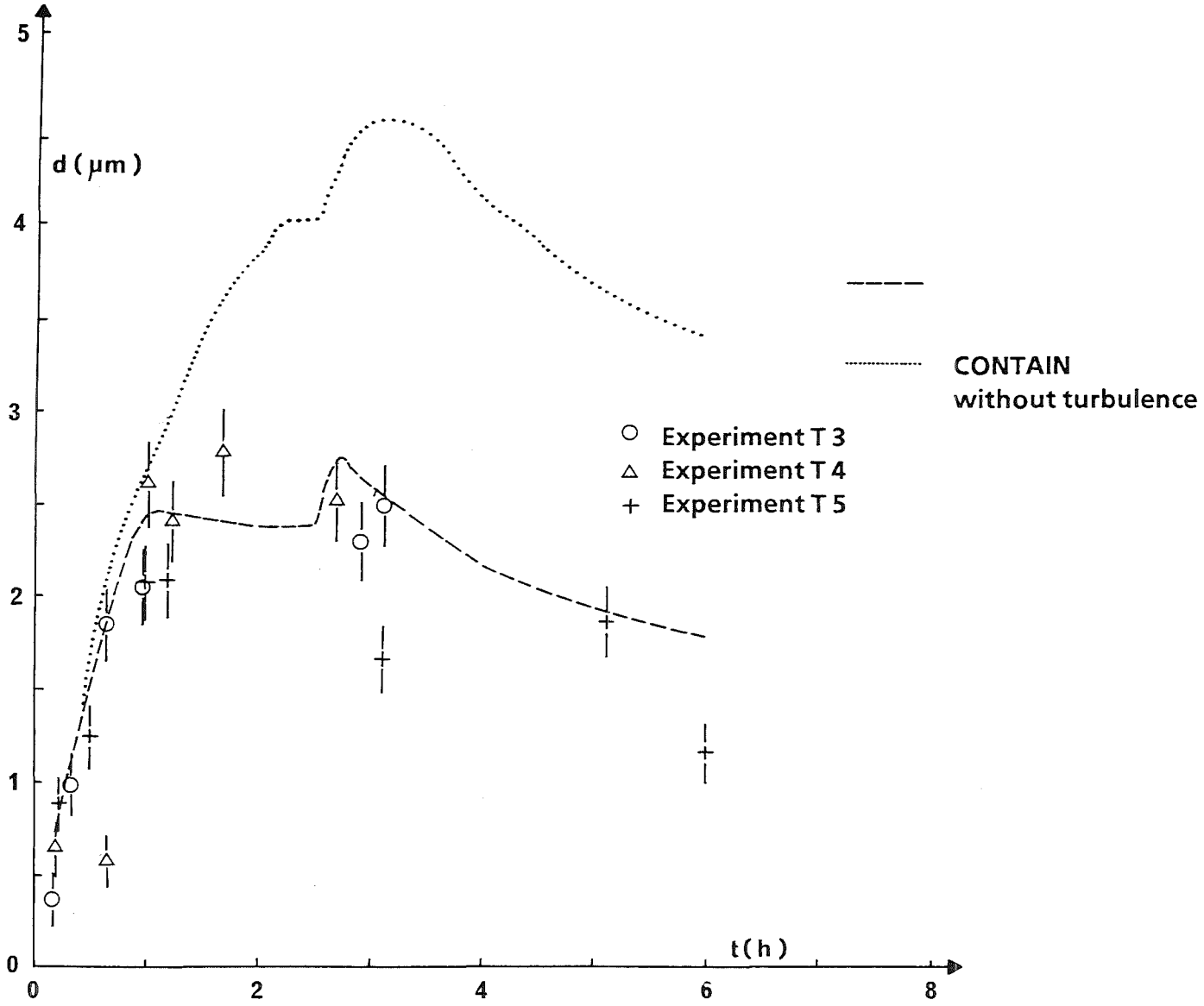

FIG. 5: COMPARISON OF THE AEROSOL-DIAMETERS MEASURED IN COURSE OF THE NALA-EXPERIMENTS TO CONTAIN CALCULATIONS
(DURATION OF SODIUM POOL HEATING: 2.5 h)

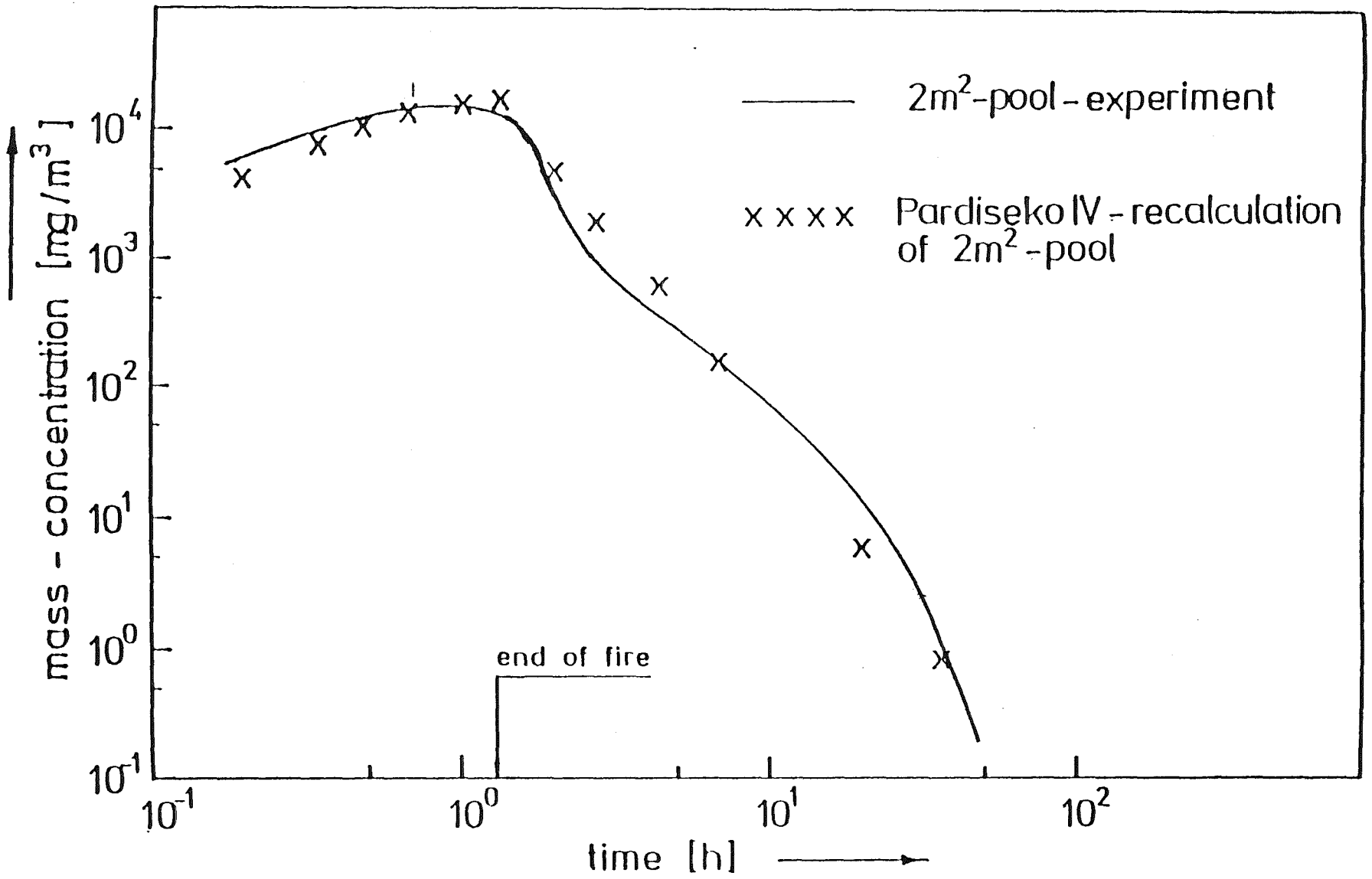

FIG. 6: COMPARISON OF THE PARDISEKO CODE TO A 2 m²-FAUNA SODIUM FIRE EXPERIMENT X = 1,1, Y = 4 (Bunz et al., 1988)

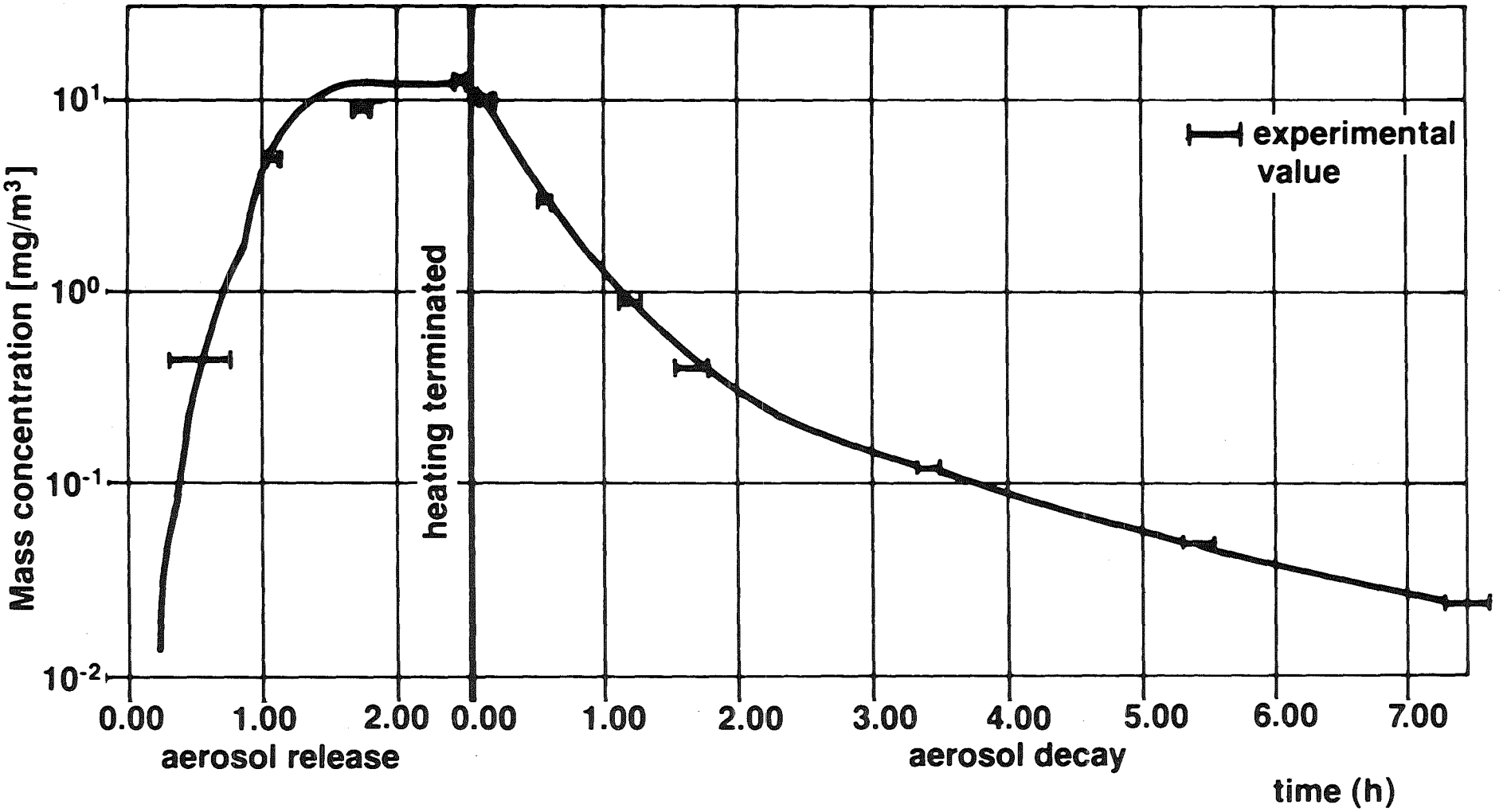

FIG. 7: PARDISEKO-COMPUTATION FOR THE AEROSOL MASS CONCENTRATION OF THE NALA SODIUM EVAPORATION EXPERIMENT T7

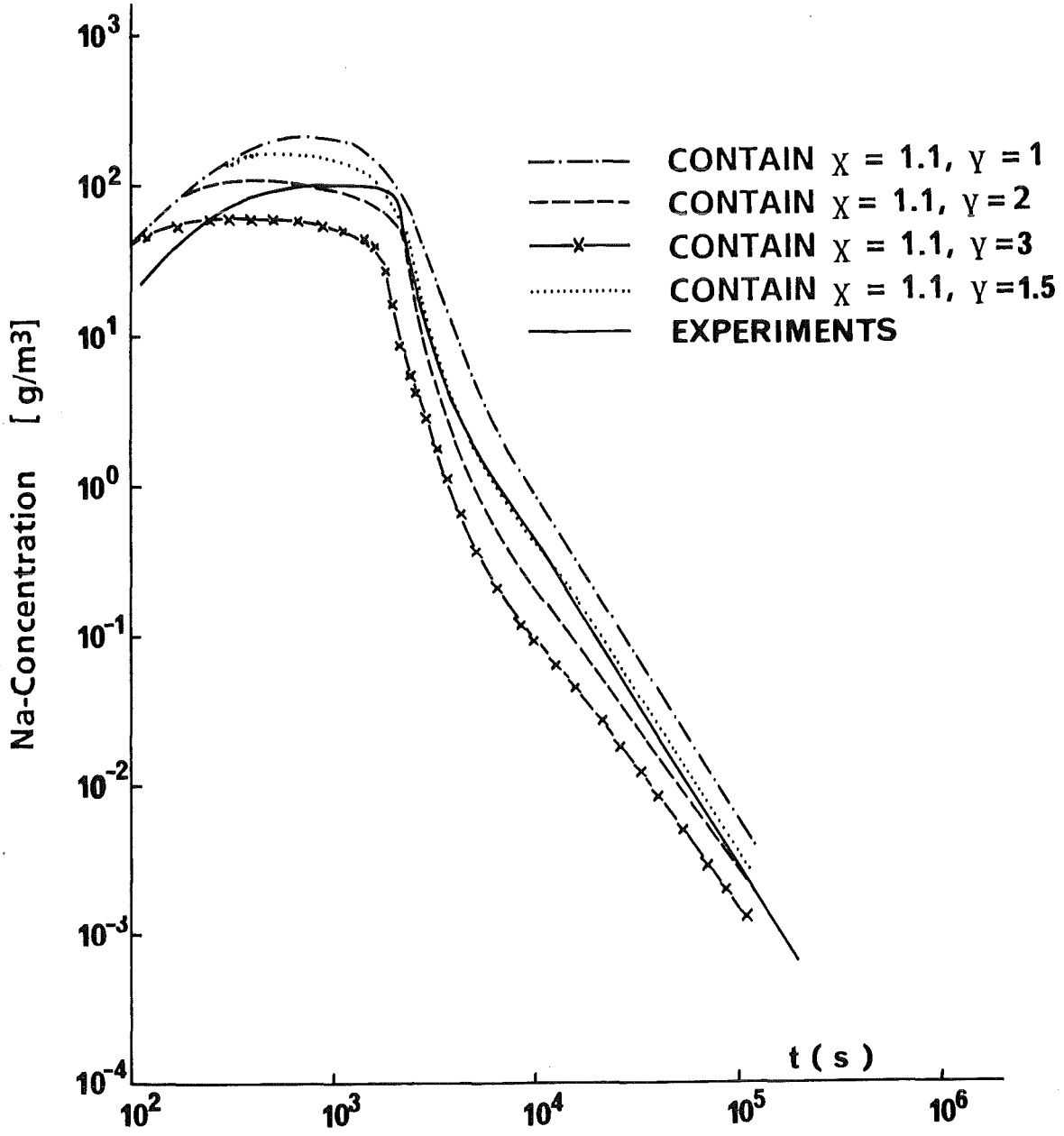

FIG. 8: SUSPENDED SODIUM MASS CONCENTRATION IN CV ATMOSPHERE (SOFICOV SA - 1)

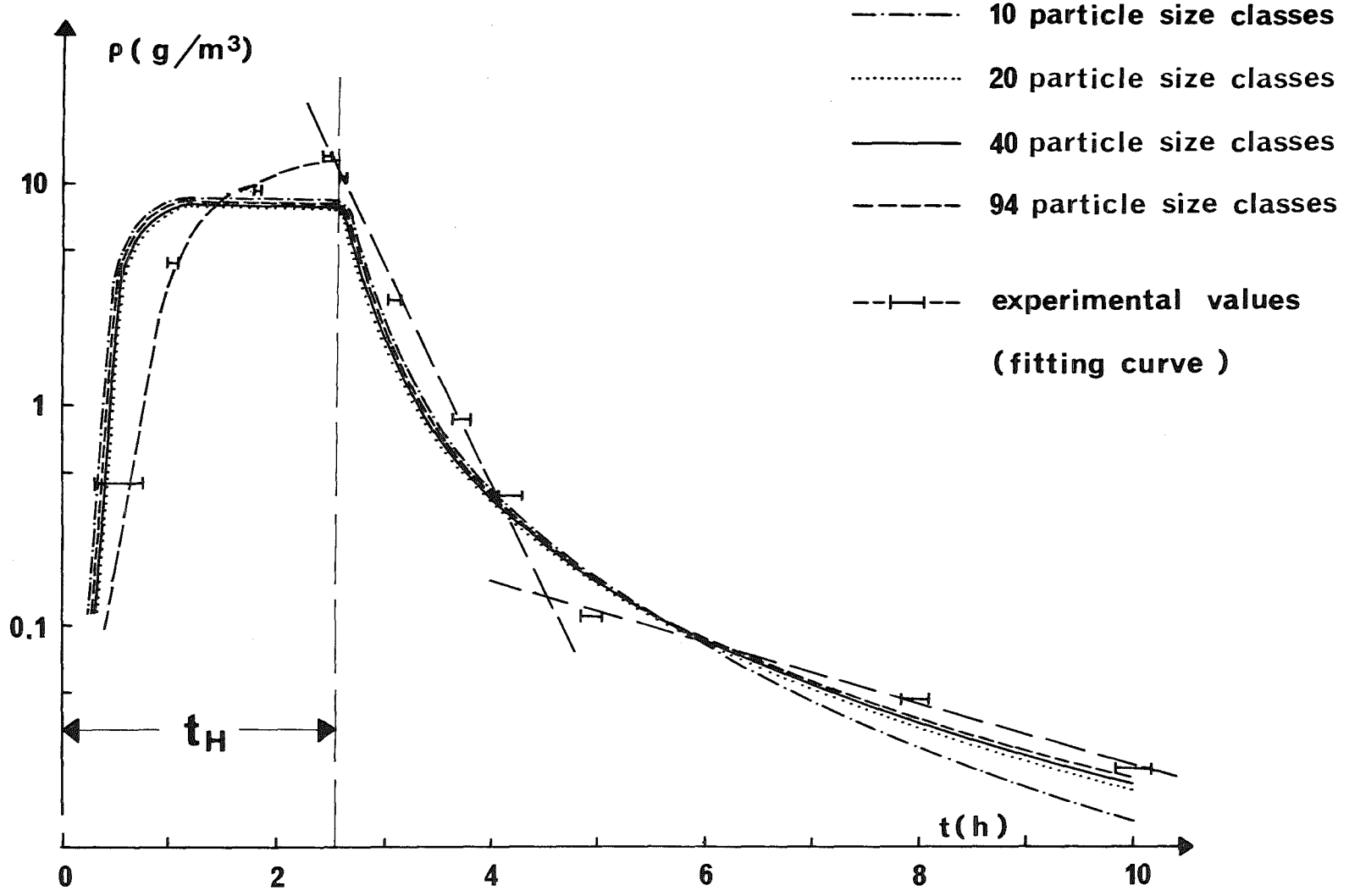

FIG.9: DEPENDENCE OF THE AEROSOL MASS CONCENTRATIONS COMPUTED WITH THE CONTAIN CODE FOR THE NALA EXPERIMENT T7 FROM THE NUMBER OF PARTICLE SIZE CLASSES