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LABORATORY DETERMINATION OF PARAMETERS FOR THE FLOW OF GAS THROUGH SOILS

S.G. IGNATIUS
Institute of Technological Research of São Paulo, São Paulo, SP, Brazil, ignatius@ipt.br

ABSTRACT: It were developed some laboratory technics for measuring the coefficients of effective intrinsic permeability, hydrodynamic dispersion and attenuation, for the flow of methane gas through a soil. It was used for the tests a "chromatograph-like" equipment that was basically an adaptation of a gas chromatograph set with a flame ionization detector, substituting the chromatographic column by a compacted soil column and analyzing the breakthrough curve with the advection-dispersion equation. The test is quick, just a few seconds in general, with the additional advantage that is not necessary to take samples periodically of the effluent to determinate the breakthrough curve, as in the traditional procedure, which eliminates the influence that it can have on the flux and also the uncertainty about the correct time interval to take samples. A sandy silt compacted in different conditions was tested by flow of methane gas. The results showed good agreement internal and with those presented in the literature.

KEYWORDS: Methane flow in soils; laboratory test; gas permeability

RESUMO: Foram desenvolvidas algumas técnicas de laboratório para medição dos coeficientes de permeabilidade intrínseca efetiva, dispersão hidrodinâmica e atenuação, para o escoamento de gás metano através de um solo. Foi utilizado nos ensaios um equipamento "tipo cromatógrafo" que era basicamente uma adaptação de um cromatógrafo a gás com detector de ionização de chama, substituindo a coluna cromatográfica por uma coluna de solo compactado e analisando a curva de chegada com a equação de advecção-dispersão. O teste é rápido, de poucos segundos em geral, com a vantagem adicional de não ser necessária coleta periódica de amostras do efluente para determinar a curva de breakthrough, como no procedimento tradicional, o que elimina a influência que pode ter no fluxo e também a incerteza sobre o intervalo de tempo correto para a coleta das amostras. Um silte arenoso compactado em diferentes condições foi testado por fluxo de gás metano. Os resultados mostraram boa concordância interna e com os apresentados na literatura.

PALAVRAS-CHAVE: Fluxo de metano em solos; ensaios de laboratório; permeabilidade ao gás

1 INTRODUCTION

Darcy's law for flow of incompressible fluids, equation (1), should be modified when applied to flow of compressible fluids like a gas, assuming the form of equation (2) for unidirectional flow of a gas (ASTM D4525-08, Bear 1988, pg. 149; Muskat 1937, pg. 78):

$$q = \frac{K_i}{\mu} \frac{(p_e - p_s)}{L} \tag{1}$$

$$q_{sg} = \frac{K_g}{\mu} \frac{(p_{ea}{}^2 - p_{sa}{}^2)}{2Lp_{sa}} \tag{2}$$

where:
q = discharge velocity, m/s;
q_{sg} = gas velocity at the exit point, m/s;
K_i = effective permeability coefficient for incompressible fluid, m^2 (=10^{12} Darcy);
K_g = gas permeability coefficient, m^2 (=10^{12} Darcy);
p_s = relative pressure of air at the exit, Pa;

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p_e = relative pressure of air at the entrance, Pa;
p_{sa} = absolute pressure of air at the exit, Pa;
p_{ea} = absolute pressure of air at the entrance, Pa;
L = length of specimen, m;
A = cross-section area of specimen, m^2; and
μ = viscosity of air at temperature of test, Pa.s.

The coefficients Ki and Kg are named "effective" because it depends on the moisture content of the soil. The values of these coefficients should equal the intrinsic permeability coefficient of the soil if the soil is saturated with the percolating fluid and the soil structure is indeformable under the efforts involved in filling it with the wetting or the non-wetting fluid.

When using the expression (2) it is necessary that the equipment be fitted with a pressure gauge to indicate the absolute pressure in the system, plus a differential pressure gauge indicator of the pressure drop along the specimen.

According to Carman (1956, pg. 62), since the experiments of Kundt and Warburg, in 1875, it is known that if the pressure in a gas flowing in a capillary is reduced until the mean free path of molecules becomes a significant fraction of the diameter of the capillary, the flow velocity exceeds the calculated by known equations for viscous flow. It is assigned to a kind of "slipping" that would occur in the contact between the gas and the solid wall, and may occur if the mean pressure, $p_m = (p_e + p_s)/2$, is too low or the soil pore diameters are too small or both. In this case the flow is no more viscous, not obeying Darcy's law, receiving the name of "Knudsen flow" in homage to one of the first researchers that described the phenomenon at the year of 1909.

The length of the average free path of gas molecules is usually in the range of about 0.01 to 0.1 µm, under normal conditions of temperature and pressure, which causes gas flow be the type of Knudsen flow in pores smaller than about 0.01 µm in diameter. Under vacuum conditions Knudsen flow can assume significance even to larger pores (Dullien 1992, PG. 289).

In a compressible flux of gas, the volumetric velocity, q, and mass density, ρ, vary as pressure, p, varies along the column flowing, in order to maintain the mass velocity, ρq, constant. As a consequence, the multiplication of p by q, pq, is also constant. Inside the column flowing, the pressure decreases from the entrance to the exit; at the exit the pressure is lesser, ρ is lesser and q is greater.

Langfelder, Chen and Justice (1968), noted that it is possible to verify the influence of consideration of compressibility of gas, by comparing the values of K and Kg obtained respectively by expressions (1) and (2). In doing this, it is possible to see that maintaining $\Delta p/pm \leq 0{,}20$ ($\Delta p = (p_s\text{-}p_e)$, e $p_m = (p_s+p_e)/2$), one should have $Kg/K \geq 0{,}90$, or, in other words, the gas permeability, and so the volumetric discharge flow velocity as well, can be determined by equation (1) for practical purposes. Without the necessity of considerate the compressibility, one can just measure the pressure drop along the specimen, avoiding the necessity of the absolute pressure determination. So, if the ambient pressure is atmospheric, it is possible the simulation of subsurface airflow using groundwater flow modeling codes, if the pressure difference between any two points in the system is less than about 0,2 atm.

Klinkenberg (1941) showed that gas permeability coefficient is inversely proportional to the mean pressure, p_m, for a single gas and is different for different gases, but tends to the intrinsic permeability coefficient as the mean pressure increases.

If two gases are present, for example methane and air, mutual mass dispersion will occur, but in general one is interested in the flux of just one of the components of the binary mixture, often methane in this case. The mass dispersion is proportional to the hydrodynamic dispersion coefficient, D, which includes diffusion, due to concentration differences and mechanical dispersion due to interaction between the gas molecules and the solid structure of the soil.

The total flux will be the composition of the advective type, and the dispersive type.

The flow has, in general, its velocity attenuated by various physical-chemical factors, usually summarized by a single factor, the so-called attenuation coefficient, R, and, since the gas can be considered

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incompressible, and the soil pores big enough to disregard Knudsen flow, can be described by the known advective-dispersive equation, that in just one dimension assumes the form:

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \frac{q}{\theta_a}\frac{\partial C}{\partial x} \tag{3}$$

Where:
R = attenuation coefficient;
D = hydrodynamic dispersion coefficient;
Θ_a = aerated porosity (air volume divided by total volume);
C = gas concentration;

The coefficients of attenuation, R, hydrodynamic dispersion, D, and permeability, K (implicit in q) are necessary inputs for the solution of the equation (3). These three coefficients can be determined in a single experiment as explained below.

2 EQUIPMENT

Tests were done to determine the three coefficients, R, D and K, for methane gas flowing through soil specimens compacted in different conditions.
The equipment used to do the tests was a chromatograph-like equipment, shown in Figure 1, constituted basically by:

- Cylinder of methane gas under pressure;
- Carrier gas (Nitrogen) cylinder;
- Methane gas pressure controller;
- Carrier gas pressure controller;
- Injector system;
- Soil specimen;
- Flame ionization detector with compressed air and hydrogen cylinders;
- Computer equipped with a software for data acquisition;

As can be seen in Figure 1, the sample gas (methane, in this case) passes through the specimen and the carrier gas by the outside, in the scheme for this study. Carrier gas collects the sample gas as soon as it passes through the output end of the specimen, and conducts it to the detector. Other configurations are possible, possibly more appropriate, depending on the case. It is possible for example to pass the carrier gas inside of the specimen, and to provide sample pulse feed of gas of interest, possibly more appropriate arrangement for testing a sample of contaminant gas from the subsoil, such as landfill gas, of which one would have only a small sample, not a bottle of compressed gas. In this case, if the gas flow inside the specimen do not have enough speed to send the sample to the detector quickly, it is possible to pass an additional current of carrier gas on the outside of the specimen to drive the effluent sample to the detector.

The distance between the exit point of the specimen and the detector should not be so large that dispersion in this way may influence the value of the coefficient of dispersion of soil that is intended to measure, nor so small that the functioning of the detector can have influence on the natural characteristics of the stream inside the specimen.

In the Figure 2 it can be seen the injector system to drive gas flux from the exit point at the soil specimen to the detector. It is constituted basically by a needle adapted to the tube at the end of the specimen that is inserted on a small chamber through a rubber sealant cork, to drive the sample beyond the point of entrance of carrier gas.

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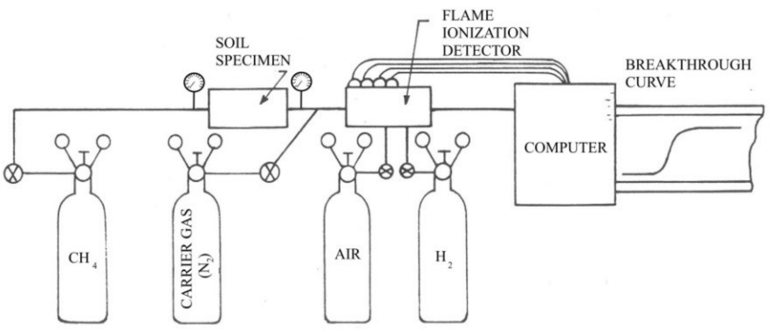

Figure 1. Equipment used in the experiments (from Ignatius, 1999)

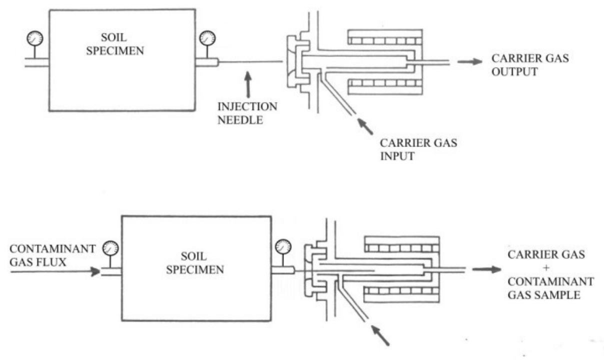

Figure 2. Injector system to drive gas flux from the exit point at the soil specimen to the detector (from Ignatius, 1999)

3 SOIL

The soil used was an elastic sandy silt with LL = 54%; LP = 32%; γ_s = 27.3 kN/m^3; $\gamma_{d\,max}$ = 15.5 kN/m^3; w_{opt} = 20%; L = 0.15 (non-lateritic, Ignatius, 1999).

Samples of the soil were compacted in various conditions of specific weight and moisture content, from which cylinders 5x5 cm were worked out and tested immediately, or dried to different moisture content before test. The tests were performed by methane flow parallel to the compaction efforts. Some results are presented in the sequence.

4 TESTS

At the top and bottom of the specimens were placed dry porous stones (low entry pressure). The compacted soil specimens were submitted to a flow of methane gas under a constant pressure difference, low enough to make the moisture content of the specimen remain constant during the test. The methane gas was injected at the entrance of the specimen in a concentration C_0 constant. The gas flowed out of the specimen directly in the stream of carrier gas and was driven to the flame ionization detector (working under the influx of air and hydrogen) that analyzed the mixture, indicating the variation of the concentrations of methane over time. The signals from the detector were sent to a computer equipped with a software that registered the shape of the breakthrough curve.

When the concentration of methane in the output equals the entry, becoming constant too, this phase of the test ended. The pressures at the entrance and at the exit point of the specimen were measured.

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Next, it was connected a bublemeter at the exit point of the specimen to measure the non-attenuated flow rate of methane through the specimen, which was saturated of methane at this time (assuming instantaneous equilibrium between masses of methane in the gaseous phases, dissolved in the liquid, and adsorbed to the solids). It was done to determine the value of the permeability coefficient, K.

Some results obtained in the tests are presented below, as examples.

5 CALCULATIONS

5.1 Permeability coefficient

The permeability coefficient, K, can be calculated directly from the measures of pressures and flow rate, by the equations (1) or (2), respectively, depending whether the flow can be considered incompressible or not.

5.2 Attenuation coefficient

The difference between a flow attenuated and not attenuated, are represented in Figures 3 and 4.

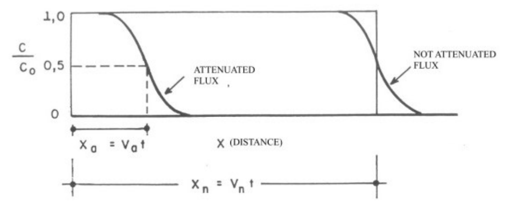

Figure 3. Comparison of concentration distributions for an attenuated and a not attenuated flow, relative to the distance travelled, x (t = time elapsed) (adap. Domenico and Schwartz 1990, pg. 640)

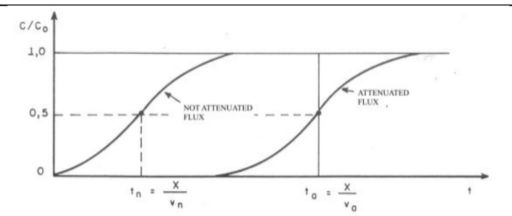

Figure 4. Comparison of concentration distributions for an attenuated and a not attenuated flow, relative to the time elapsed, t (x = distance travelled) (adap. Domenico and Schwartz 1990, pg. 640)

The effect of the attenuation process is the removal of part of the mass of the fluid that moves, at the front of spread, reducing the concentration and therefore reducing the mass flow velocity. The velocity $v_n = q_n/\theta_a$ is the average interstitial velocity of the stream without attenuation and without dispersion, and v_a is the interstitial attenuated velocity also without dispersion (speed of point $C/C_0 = 0.5$). The attenuation coefficient, often symbolized by the letter R (for "Retardation") is defined as the ratio of v_n and v_a, namely:

$$R = \frac{v_n}{v_a} \tag{4}$$

The coefficient R, when applied to the point $C/C_o = 0.5$, reduces the speed of this point and also of all the other points of the curve of concentration, so that the curve attenuated in Figure 4 is obtained by a simple translation of the curve not attenuated, to a value of R, i.e. the length actually travelled x_a, equals x_n/R.

As $t_n = x/v_n$ and $t_a = x/v_a$, x being the specimen length, it can be seen, from equation (4), that:

$$R = \frac{t_a}{t_n} \tag{5}$$

In the case of an incompressible fluid flow through a body of length x = L, determination of R through the expression (5) is straightforward: t_a is determined directly in the test, for C/Co = 0.5, and t_n is calculated from v_n that is constant along the stream and calculated from expression (1) divided by θ_a.

However, in the case of a compressible gas flow through a body of length x = L, v_n is variable along the stream. The expression for t_n must be obtained by integration:

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$$t_n = \int_0^L \frac{dx}{v_n} \tag{6}$$

Effecting this integration results (Ignatius, 1999):

$$t_n = \frac{4\theta_a \mu L^2 (p_e^3 - p_s^3)}{3K(p_e^2 - p_s^2)^2} \tag{7}$$

Having obtained t_n by expression (7) and determined t_a directly from the test, the value of R is calculated through the expression (5).

5.3 Hydrodynamic dispersion coefficient

The attenuated advective-dispersive movement of a gas through a porous medium can be represented by the equation (8) which is the transcript of the equation (3), where instead of q is written q_n, to emphasize that it is the discharge velocity of the fluid in question, without attenuation:

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \frac{q_n}{\theta_a}\frac{\partial C}{\partial x} \tag{8}$$

A short solution to the equation (8) is, in a test in which one imposes the conditions:

$C(0,t) = C_0$,

$C(x,0) = 0$,

$$C(L,t) = \left(\frac{C_0}{2}\right)\mathrm{erfc}\left[\frac{(RL - v_n t_a)}{2(DRt_a)^{1/2}}\right] \tag{9}$$

In equation (9) erfc is the complementary error function, which occurs frequently in solutions of the equation (9). This is a well-known function, for which charts and graphs can provide numeric values (for ex. in Domenico and Schwartz, 1990, PG. 637). In this equation, L is the length of the specimen and C is the concentration corresponding to the time t_a, experimentally determined at the output of the specimen.

In the case of an incompressible fluid flow, the value of v_n, which is constant along the stream, could be easily calculated from expression (1) divided by θ_a.

In the case of a compressible gas flow through a body of length x = L, v_n is variable along the stream. It is possible, however, best approximate the physical phenomenon using in the equation (9) a hypothetical value of equivalent constant velocity, $v_{n\,eq}$, which would be the one that would cause a gas particle traverse the length L at time t_n (given by equation (7)). The value of the velocity equivalent $v_{n\,eq}$ can then be calculated using the expression:

$$v_{n\,eq} = \frac{L}{t_n} \tag{10}$$

The value of t_n is calculated by the expression (7). The expression (9) so becomes:

$$\frac{C(L,t)}{C_0} = \frac{1}{2}\mathrm{erfc}\left[\frac{(RL - v_{n\,eq} t_a)}{2(RDt_a)^{1/2}}\right] \tag{11}$$

The value of the hydrodynamic dispersion coefficient, D, is finally determined adjusting the theoretical curve expressed by (11) to the experimental curve $C/C_0 = f(t_a)$ for the best value of D.

6 RESULTS OBTAINED

Some results of effective air permeability coefficients are shown in the Figure 5, where it can be seen that they agree well with the expectations.

Figure 6 show results for attenuation coefficient, R, varying with moisture content (specimens compacted and tested at the same moisture content each) for two different values of dry density (1450 and

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1550 kg/m^3). It can be seen that, even the solubility of methane in water being low, R increases with moisture content.

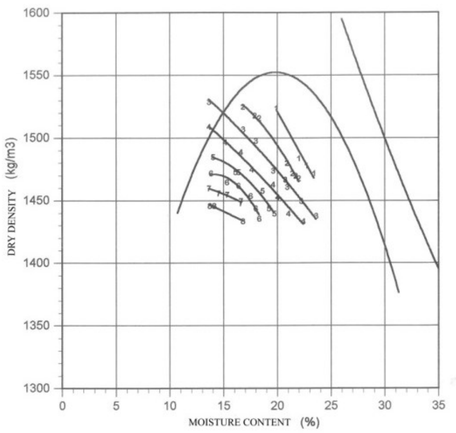

Figure 5. Isovalue curves of effective air coefficient of permeability, K_a, related to the compaction curve and the 100% water saturation curve. Numbers in the figure (1 to 8) are the values of $K_a/10^{-14}$ m^2 (from Ignatius, 1999)

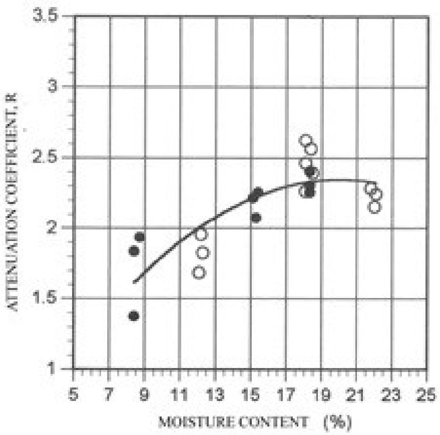

Figure 6. Attenuation coefficient, R, varying with moisture content (specimens compacted and tested at the same moisture content each) for two different values of dry density (1450 and 1550 kg/m^3) (adap. Ignatius, 1999)

Figure 7 presents the experimental and the adjusted breakthrough curves for one of the tests and the value of D obtained.

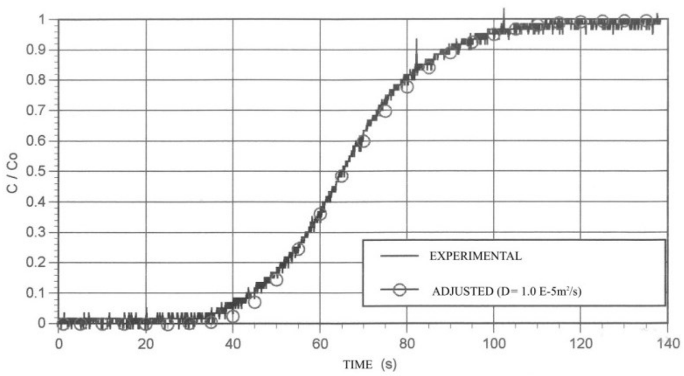

Figure 7. One test result as an example –experimental and adjusted breakthrough curves for determination of hydrodynamic dispersion coefficient, D (from Ignatius, 1999)

As can be seen in the Figure 7, the test is quick, just a few seconds in general (less than 140 seconds in this case), with the additional advantage that is not necessary to take samples periodically of the effluent to determinate the breakthrough curve, as in the traditional procedure, which eliminates the influence that it can have on the flux and the uncertainty about the correct time interval to take samples.

Values of D for different values of equivalent constant advective velocity, $v_{n\ eq}$, for specimens submitted to different compaction conditions but same aerated porosity, θ_a, are shown in Figure 8. It can be seen in the figure that is possible to estimate the value of D for zero advective velocity, D_0, extrapolating the results. In the Figure 9, the results obtained are compared with some results from the literature showing same tendency and relative good agreement, as can be seen. The relation D_0/D_f, where D_f, is free diffusion coefficient of methane in air and D_0 is the mean value, for each θ_a, of the value of D extrapolated for zero advective velocity, is presented against aerated porosity, θ_a.

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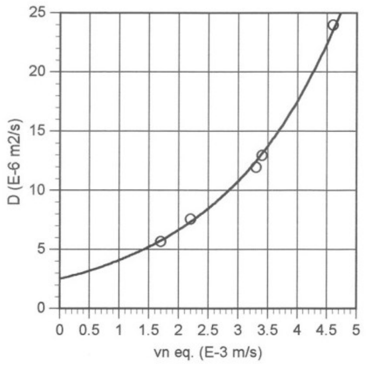

Figura 8. Hydrodynamic dispersion coefficient in function of advective flow velocity ($\theta_a = 0.15$) (from Ignatius, 1999)

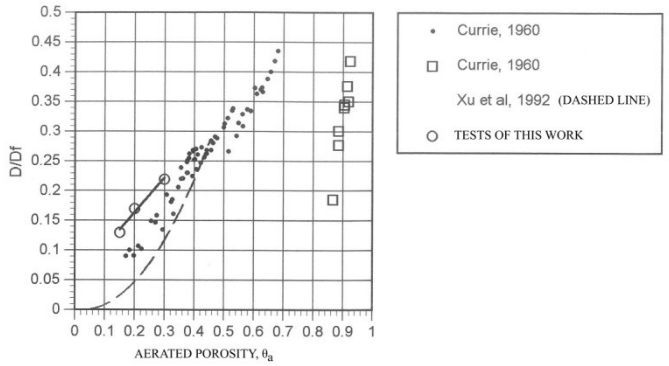

Figura 9. Relation between D_0 (for zero advective flow velocity) e D_f in function of θa para alguns tipos de materiais (tests of this work and results from Currie 1960 and Xu, Nieber e Gupta 1992). (Currie at left: glass spheres, sand, NaCl, carborundum, soil aggregates; Currie at right: mica; Xu Nieber e Gupta 1992: two silty clays – one illitic and other montmorillonitic and two clayey fine sand (from Ignatius, 1999)

7 CONCLUSIONS

It is presented in this paper a method to obtain experimentally the permeability, attenuation and hydrodynamic dispersion coefficients for methane gas flow through compacted soil samples in a single test. Some results are presented that showed good agreement with expectations and with the literature.

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