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Title: Geochemical Natural Analog Research — Model Development

Names of individuals performing the activity:

Bill Murphy [signature]
Sitakanta Mohanty
Ashok Nedungadi

Objective: The objective can be referred to in Section 3.1.4 "Task 4: Interpretation of data and modeling" of the project plan titled "Geochemical Analog of Contaminant Transport in Unsaturated Rock", Rev 2 Chang 7, August 1994, Pg 15-16.

Objectives to be accomplished:
Develop conceptual models of contaminant transport at the sites. Use quantitative models to evaluate contaminant transport and other events and processes that occurred during the evolution of the natural analog site.

Summary of technical approach to be used:
Establish a conceptual model based on available data. The model should be simple enough for quantitative interpretation using established codes (numerical).

Modeling may be done in advance of data collection, in order to test predictive capability. Site data will be used to constrain the initial and boundary conditions for the modeling.

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Results of model prediction will be compared with the site data. This may lead to improvements in the models and identification of additional site or generic data required to support the models. Based on the success of the models for interpretation, the usefulness of the natural analog data for model validation will be assessed.

Modeling work to be documented here pertains to the Akrotiri site in Greece. The hydrologic data collected from this site will be used in assessing the effects of intermittent water infiltration over 3600 years of burial period. The boundary conditions for the model will be based on field and laboratory measurements and observations.

PORFLOW code will be used in this excercise to develop initial flow models for Akrotiri site.

Identification:

Configuration management status:

Computer platform used : unix workstation, a SPARC 10 computer

Directory and files are currently residing in DOPEY at

bmurphy/porflow

The main program and the peripheral programs are written in FORTRAN 77 language and

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are compiled by using f77 option.

The computational reliability can be affected by the control parameters which are standard to all flow calculations such as grid size, timesteps etc.

A dataset for PORFLOW representing the conceptual model is presented in pages 4–9.

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akrotiri.~~txt~~ 11

```
/----------------------------------------------------------------------
/ AKROTIRI problem: unsaturated flow and transport
/----------------------------------------------------------------------
/
GRID 601 by 3            (x,y)   $$$$$
COORdinate X CART MINImum=0.0, MAXImum=24.0 meters
COORdinate Y CART -1.0 ,  0.0,  1.0  meters
/
DATUM 0, 0 (at the surface)
/
GRAVITY is 9.81, 0.                        $ flow along positive X direction
/
Density for primary fluid: uniform at 1000. kg_per_m cubed
/
/----------------------------------------------------------------------
ZONE 1                   FROM (1,1) TO (38,3)     $$$$$  Minoan
FOR                      zone 1
ROCK                     density = 2130, eff = 0.5, tot = 0.5, diff = 0.5
HYDRaulic props for P    ss = 1.e-6, (Kx,Ky) = 2*1.75e-4 (m/s)
MULTiphase               VAN  n= 2.000  alpha= 0.1570  swr= 0.25
/ Kd is in m3/kg  and molecular diffusivity is in m2/sec
Transport for C:  kd=0.0027, dm=8.0e-10, Ld = 0.004 , Td = 0.0004
MATE TORT 0.5 0.5
/----------------------------------------------------------------------
ZONE 2                   FROM (39,1) TO (39,3)   $$$$$  Packed Earth
FOR                      zone 2
ROCK                     density = 2130, eff = 0.3, tot = 0.3, diff = 0.3
HYDRaulic props for P    ss = 1.e-6, (Kx,Ky) = 2*1.00e-7 (m/s)
MULTiphase               VAN  n=1.131   alpha= 2.700  swr= 0.385
/ Kd is in m3/kg  and molecular diffusivity is in m2/sec
Transport for C: kd=0.45, dm=8.0e-10, Ld = 0.004 , Td = 0.0004
MATE TORT 0.15 0.15
/----------------------------------------------------------------------
ZONE 3                   FROM (40,1) TO (601,3)   $$$$$  Cape Riva
FOR                      zone 3
ROCK                     density = 2130, eff = 0.5, tot = 0.5, diff = 0.5
HYDRaulic props for P    ss = 1.e-6, (Kx,Ky) = 2*1.5e-4 (m/s)
MULTiphase               VAN  n=2.0   alpha= 0.07885  swr= 0.02
/ Kd is in m3/kg  and molecular diffusivity is in m2/sec
Transport for C: kd=0.0027, dm=8.0e-10, Ld = 0.004 , Td = 0.0004
MATE TORT 0.25 0.25
/
/----------------------------------------------------------------------
/
LOCATE (26,1) to (38,3)                    $ represents the contaminant source
/SOURCE C is constant at 0.8E-4 VOLUmetric kg/sec/cu m in SELEcted region
/solubility limits are in gm/liter=kg/m3
SOURCE C SOLUbility limited Cs= 6.3546e-4, S = 100 kg, t=0 SELEcted region
/
PROPerty GEOMetric
/
INTEgration for C by CONDIF
/BOUNdary for P  at -1 FLUX  is 1.E-19 (m/s) $fixed flow rate
/BOUNdary for P  at -1 FLUX  is 6.4300411e-8 (m/s) $fixed flow rate
/BOUNdary for P at -1 FLUX = 3.1709791e-9 * SIN ( 1.9923849908611e-7 * TIME
/   + 0. ) + 0.
/
BOUNdary for P at -1 FLUX = 9.96192e-9 * SIN ( 1.992384991e-7 * TIME + 0.)
 + 4.7564688e-9
/Boundary for P  at -1 FLUX  is 1.595490e-9 (m/s) $fixed flow rate
```

```
BOUNdary for P at +1 value  0.0  m $head zero at water table
Boundary for P  at -2 FLUX  is 0.0    $nothing flows in along y direction
Boundary for P  at +2 FLUX  is 0.0    $nothing flows out along y direction
/
Boundary for C   at -1: GRAD  =  0.
/Boundary for C   at +1: GRAD  =  0.
Boundary for C   at -2: FLUX  =  0.
Boundary for C   at +2: FLUX  =  0.
/
INITIAL C  is 1.e-8 everywhere
/
/INITIAL P is -12.0 from (1,1) to (601,3)    $$$$$$
/SET LINE P = -24 +1 * X
/SET LINE P = -12.
/INIT U = 1.e-15
INITial P is  0.000000000E+00 from   601    1 to   601    3
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INITial P is -0.221065283E+02 from    48   1 to    48   3
INITial P is -0.221464273E+02 from    47   1 to    47   3
INITial P is -0.221863257E+02 from    46   1 to    46   3
INITial P is -0.222262235E+02 from    45   1 to    45   3
INITial P is -0.222661206E+02 from    44   1 to    44   3
INITial P is -0.223060170E+02 from    43   1 to    43   3
INITial P is -0.223459127E+02 from    42   1 to    42   3
INITial P is -0.223858078E+02 from    41   1 to    41   3
INITial P is -0.224257021E+02 from    40   1 to    40   3
INITial P is -0.224655958E+02 from    39   1 to    39   3
INITial P is -0.225054888E+02 from    38   1 to    38   3
INITial P is -0.225453811E+02 from    37   1 to    37   3
INITial P is -0.225852727E+02 from    36   1 to    36   3
INITial P is -0.226251637E+02 from    35   1 to    35   3
INITial P is -0.226650539E+02 from    34   1 to    34   3
INITial P is -0.227049434E+02 from    33   1 to    33   3
INITial P is -0.227448322E+02 from    32   1 to    32   3
INITial P is -0.227847203E+02 from    31   1 to    31   3
INITial P is -0.228246076E+02 from    30   1 to    30   3
INITial P is -0.228644943E+02 from    29   1 to    29   3
INITial P is -0.229043802E+02 from    28   1 to    28   3
INITial P is -0.229442654E+02 from    27   1 to    27   3
INITial P is -0.229841498E+02 from    26   1 to    26   3
INITial P is -0.230240335E+02 from    25   1 to    25   3
INITial P is -0.230639165E+02 from    24   1 to    24   3
INITial P is -0.231037987E+02 from    23   1 to    23   3
INITial P is -0.231436802E+02 from    22   1 to    22   3
INITial P is -0.231835610E+02 from    21   1 to    21   3
INITial P is -0.232234409E+02 from    20   1 to    20   3
INITial P is -0.232633201E+02 from    19   1 to    19   3
INITial P is -0.233031986E+02 from    18   1 to    18   3
INITial P is -0.233430763E+02 from    17   1 to    17   3
INITial P is -0.233829532E+02 from    16   1 to    16   3
INITial P is -0.234228293E+02 from    15   1 to    15   3
INITial P is -0.234627047E+02 from    14   1 to    14   3
INITial P is -0.235025792E+02 from    13   1 to    13   3
INITial P is -0.235424530E+02 from    12   1 to    12   3
INITial P is -0.235823260E+02 from    11   1 to    11   3
INITial P is -0.236221982E+02 from    10   1 to    10   3
INITial P is -0.236620696E+02 from     9   1 to     9   3
INITial P is -0.237019401E+02 from     8   1 to     8   3
INITial P is -0.237418099E+02 from     7   1 to     7   3
INITial P is -0.237816789E+02 from     6   1 to     6   3
INITial P is -0.238215470E+02 from     5   1 to     5   3
INITial P is -0.238614143E+02 from     4   1 to     4   3
INITial P is -0.239012808E+02 from     3   1 to     3   3
INITial P is -0.239411465E+02 from     2   1 to     2   3
INITial P is -0.239810113E+02 from     1   1 to     1   3
CONV P  LOCAl criterion=1.E-5, max iterations 100
//CONV P,      1.E-3, 1
//CONV P2,     1.E-3, 1
//CONV flow,   1.E-3, 5
/
/Matrix sweeps in X direction for P=1 use ADI method
//Matrix p=2, T=3
DIAGNOSTIC S, C for NODE at (2,2) EVERY 100 steps    $$$$$
RELAXation factor for P, C 0.99
/
HISTory  at (2,2)
HISTory   P, C, S every 100 steps
/
/SOLV off
/SOLVE for U, P, C  S UNTIL TIME = 1.135296e11 IN STEPS OF 100 fact
/OUTPUT P, U, S, C in XY NOW
/SAVE   P, U, S, C NOW
/end
/quit
/OUTPUT P, U, S, C in XY NOW
/SAVE   P, U, S, C NOW
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/211.4 days=1.826496e7 sec
/33608 days = 2.9037312e07 sec
SOLVE U, P, C, S 1.826496e7 IN STEPS OF 1440 fact 1.005 max 86400
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/ten years
SOLVE U, P, C, S 3.1536e8  more IN STEPS OF 259200 fact 1.01 max 518
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/one hundred years
SOLVE U, P, C, S 2.83824e9  more IN STEPS OF 518400 fact 1.01 max 86
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/one thousand years
SOLVE U, P, C, S 2.83824e10  more IN STEPS OF 864000 fact 1.01 max 1
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW


/
/two thousand years
SOLVE U, P, C, S 3.1536E10  more IN STEPS OF 1728000 fact 1.01 max 172
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/three thousand years
SOLVE U, P, C, S 3.1536e10  more IN STEPS OF 1728000 fact 1.01 max 172
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/thirtysix hundred years
SOLVE U, P, C, S 1.89216e10  more IN STEPS OF 1728000 fact 1.01 max 17
OUTPUT P, U, S, C in XY NOW
SAVE   P, U, S, C NOW
/
/OUTPUT u,v,p,p2, T, S, c , rho, rho2, rho3, s3, eqva in NARROW format
/save OFF
end
```

6/6/95

Schematic of the conceptual model

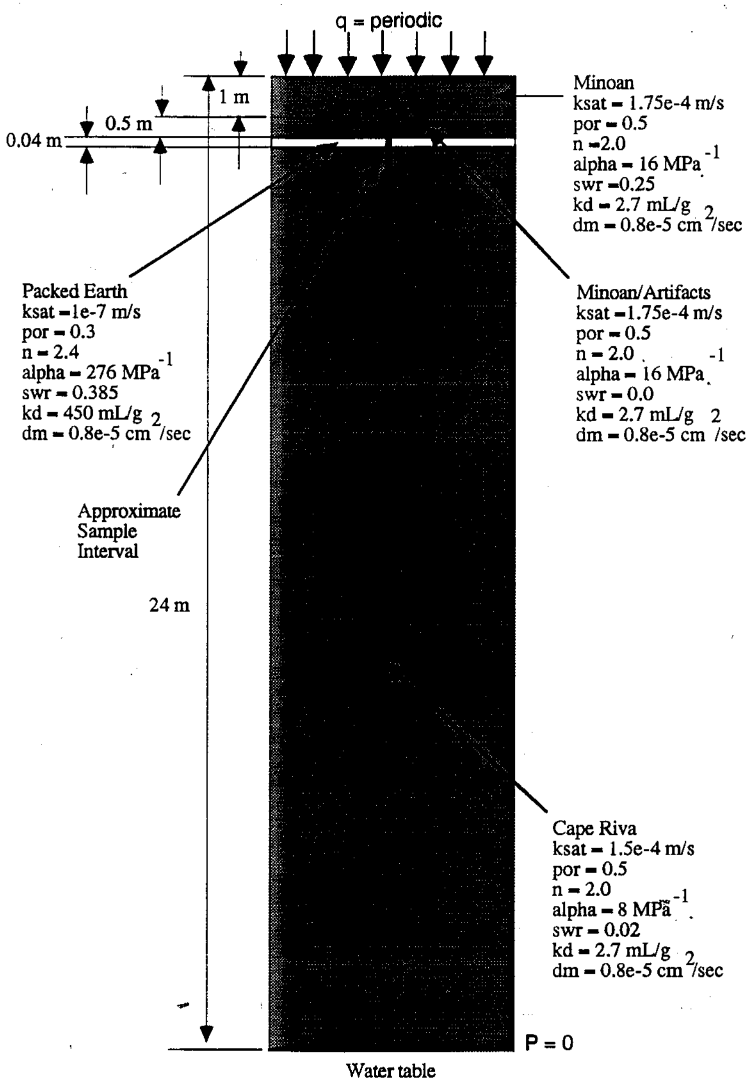


Figure 4-8. Schematic of 1D model geometry and model parameters. Symbolically, ksat represents the saturated hydraulic conductivity, por stands for porosity, n and alpha are Van Genuchten parameters, swr represents the residual water content, kd stands for the solid-liquid distribution coefficient for Cu (K_d), and dm stands for the molecular diffusivity of aqueous Cu. Figure is to scale except exaggeration of the packed earth level by 6.5×.

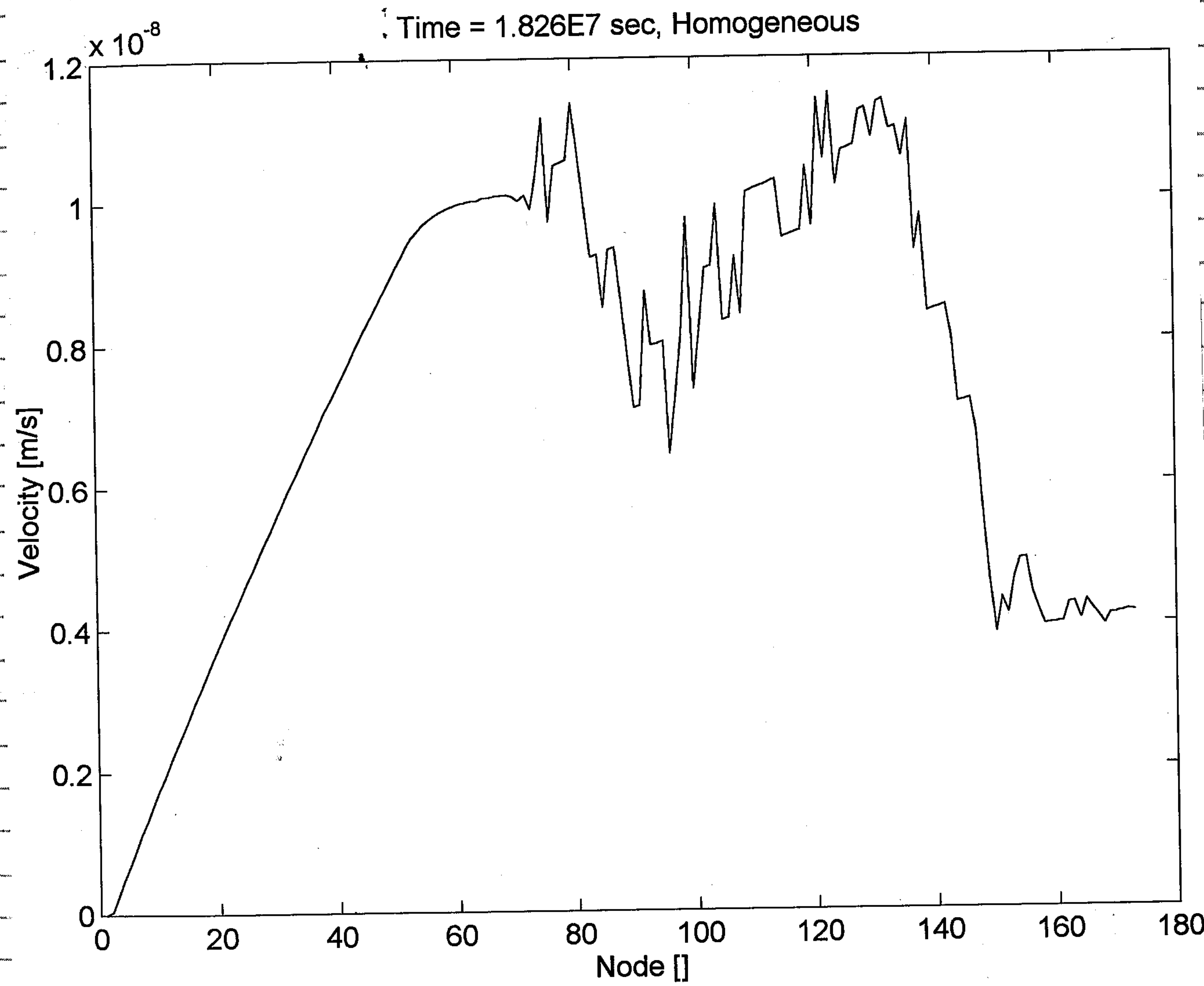
6/6/95

04/1/95

RAN PORFLOW ON THE INPUT FILE AKROTIRI·TXT11

OBSERVED FLUCTUATIONS IN THE VELOCITY PLOTS (SEE FIGURES BELOW)

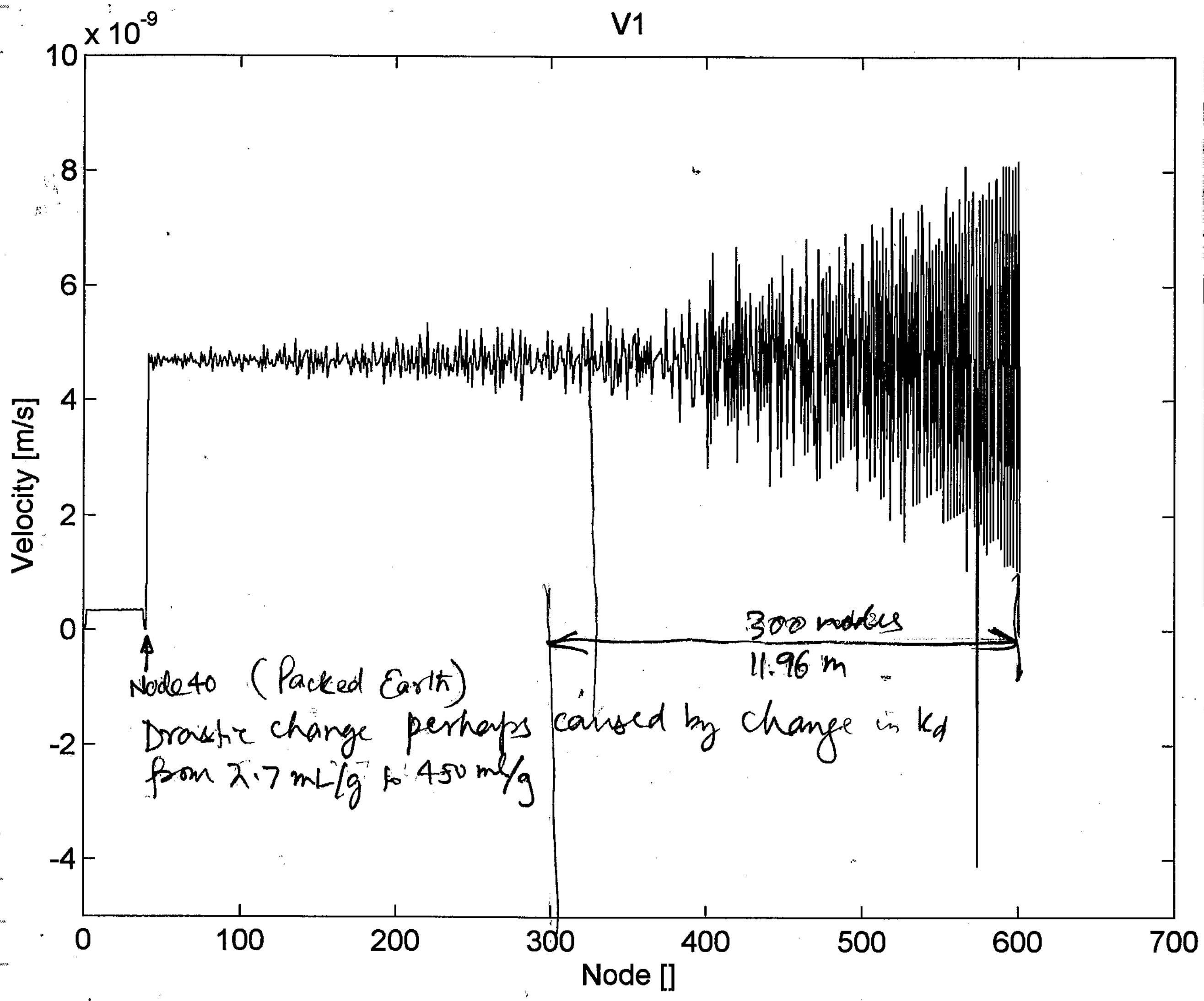
UPON HOMOGENIZING 1D DOMAIN



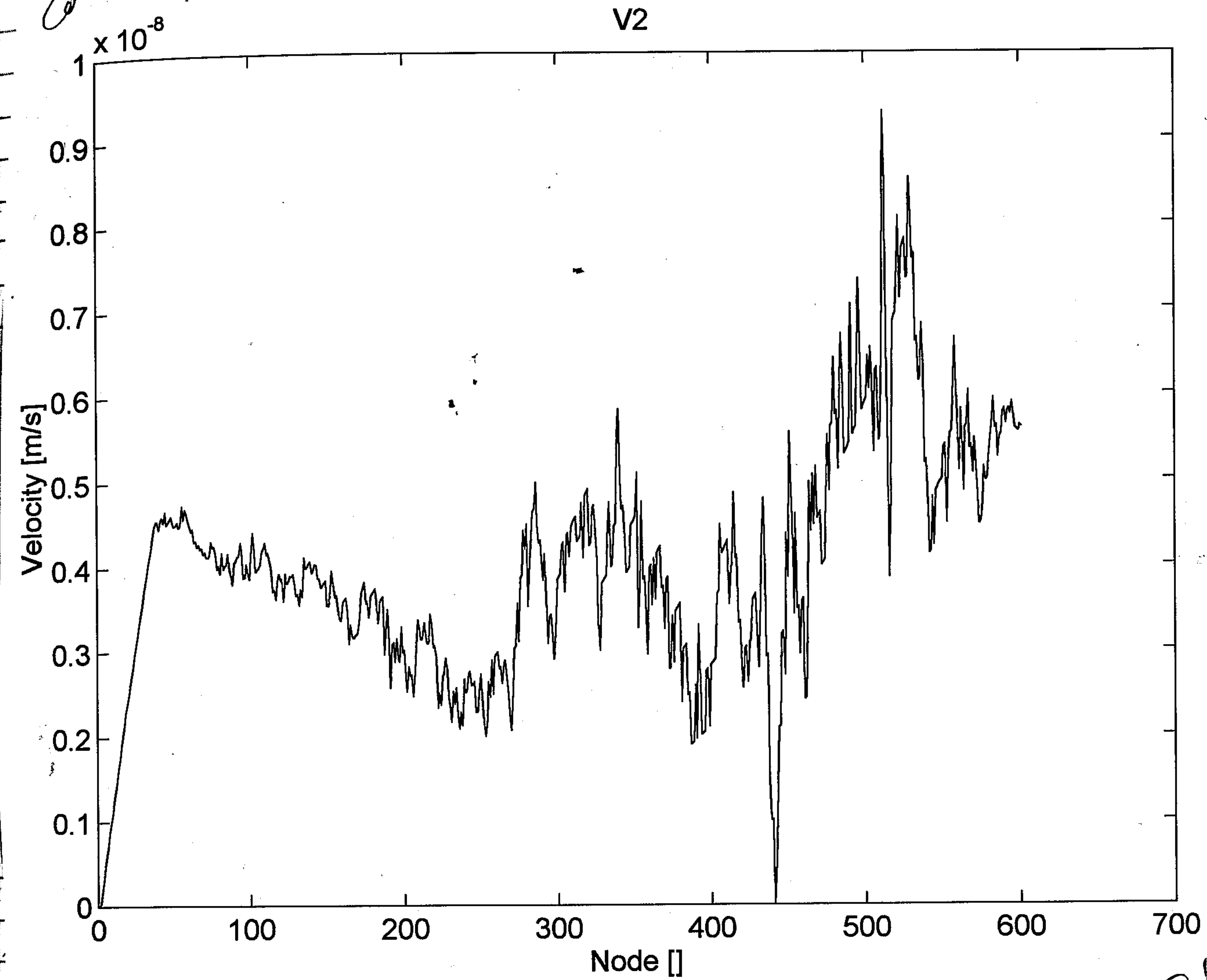
6/6/95

04/04/95

TRIED THE SAME INPUT FILE AKROTIRI.TXT ~~ON~~ WITH THE FULL HETEROGENEOUS DOMAIN. SAME FLUCTUATIONS OBSERVED IN VELOCITY PROFILE. SEE FIGURE BELOW



AWM 6/6/95



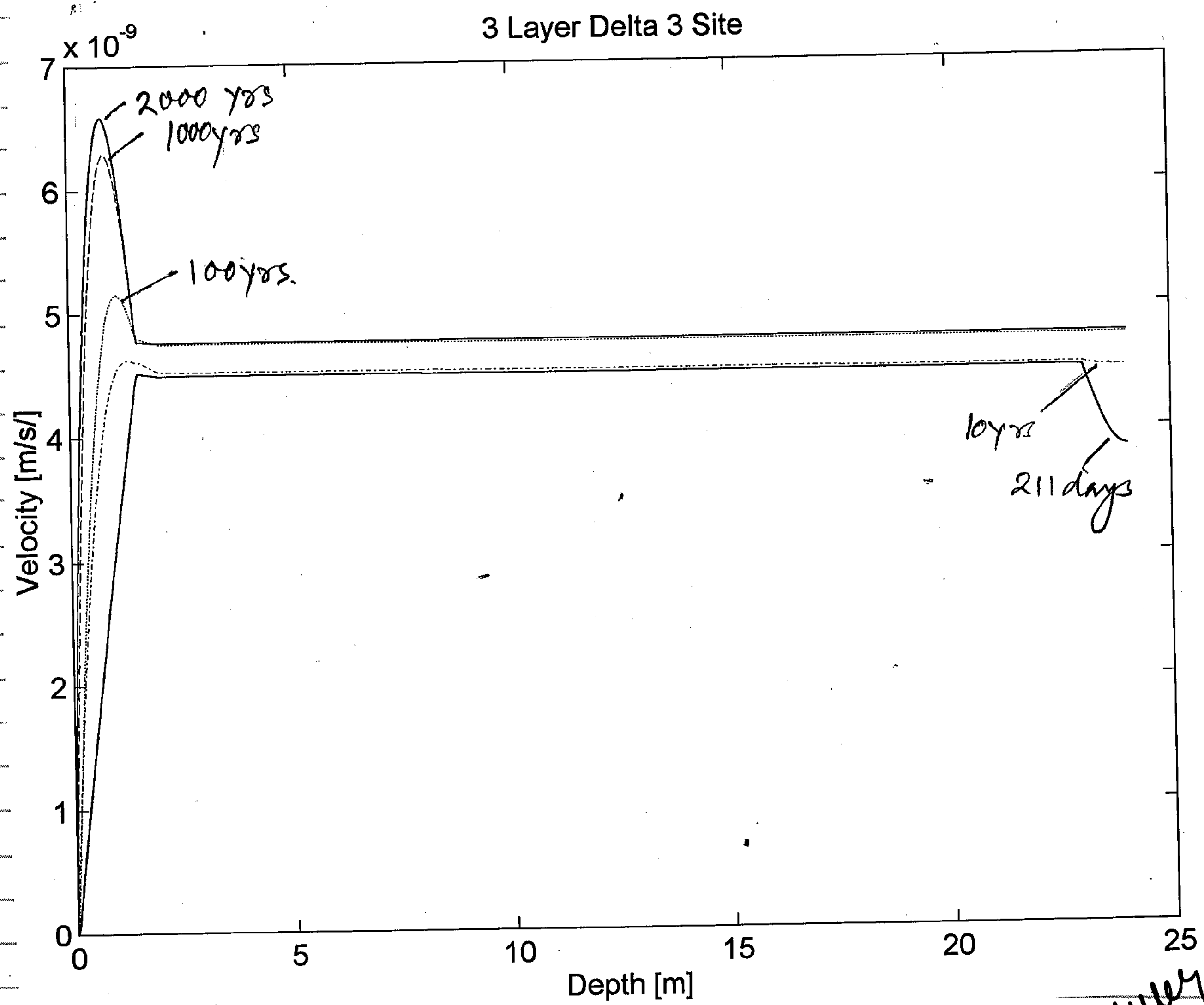
REFINED GRID → NO CHANGE IN FLUCTUATIONS OF VELOCITY PROFILE

AWM 6/6/95

04/07/95

IN ORDER TO ATTEMPT TO REMOVE FLUCTUATIONS IN VELOCITY PROFILE, I RECOMPILED PORFLOW USING THE "-r8" OPTION ⇒ DOUBLE PRECISION VERSION OF PORFLOW.

RAN THE DOUBLE PRECISION VERSION OF PORFLOW ON AKROTIRITXT11. OBSERVED THAT THE FLUCTUATIONS DISAPPERED. SEE FIGURE BELOW.



QW WMM 6/6/95

04/10/95

CHECKED SATURATION & CONCENTRATION PROFILES USING DOUBLE PRECISION PORFLOW AND PREVIOUS RESULTS. SATURATIONS/CONCENTRATIONS AND PRESSURE HEAD CHECKED OK. WE NOW HAVE SMOOTH VELOCITIES AS WELL.

QW

WMM 6/6/95

04/14/95

SENSITIVITY ANALYSIS :

~~CREATED THE FOLLOWING DIRECTORIES ON "DOPEY":~~ CW

OBSERVED THE EFFECT OF CHANGING :

- KSAT OF THE PACKED EARTH
- VANGENUCHTEN α OF PACKED EARTH
- θ_s " " "
- θ_r " " "
- POROSITY " " "

VARIED EACH OF THE ABOVE PARAMETERS WHILE HOLDING THE OTHERS TO THEIR ORIGINAL VALUES.

PLOTS WERE FAXED/SENT ORIGINALS TO SITAKANTA

CW

6/6/95

04/17/95

CONTINUED SENSITIVITY ANALYSIS

OBSERVED THE EFFECTS OF CHANGING

- KSAT
- α
- θ_s
- θ_r
- POROSITY

} OF THE TOP MOST LYR (MINOAN)

VARIED EACH OF THE ABOVE PARAMETERS WHILE HOLDING ALL OTHER PARAMETER FIXED (ORIGINAL VALUES AS FOUND IN "AKROTIRI.TXT").

PLOTS FAXED TO SITA KANTA.
ORIGINALS GIVEN TO SITA KANTA.

CW

6/6/95

04/24/95

SENSITIVITY ANALYSIS CONTINUED

OBSERVED THE EFFECTS OF CHANGING ~~BOTH~~ PROPERTIES OF BOTH TOP MOST & BOTTOM MOST LAYER SIMULTANEOUSLY

GN

THE FOLLOWING PARAMETERS OF THE TOP MOST (MINUAN) & BOTTOM MOST (CAPE RIVA) WERE CHANGED SIMULATANEOUSLY

- KSAT
- α
- θ_s
- θ_r
- n

PLOTS (ORIGINALS) WERE SENT TO SITAKANTA

GN

WW 6/6/95

04/28/95 GN

WROTE TEXT DESCRIBING RESULTS OF ALL SENSITIVITY ANALYSIS EXPTS PERFORMED DURING THE COURSE OF THIS MONTH.

TEXT PREPARED IN WORD PERFECT & EMAILED TO SITA KANTA.

GN 05/02/95

EDITED THE TEXT/WRITEUP BASED ON SITAKANTA'S COMMENTS.

TEXT EMAILED TO SITAKANTA FOR HIS REVIEW

GN

WW 6/6/95

05/05/95

PERFORMED A FINAL SENSITIVITY ANALYSIS ON :

INFILTERATION.

OBSERVED THE EFFECT OF VARYING THE INFILTERATION ON THE CONCENTRATION FRONT @ 211 Days, 10 yrs, 100 yrs, 1000yrs, 2000 yrs.

THESE RESULTS WERE PLOTTED & SENT TO SITAKANTA

AN

6/6/95

05/12/95

FINAL ARCHIVING OF ALL DATA FILES. REQUESTED ALFRED JOHNSON TO ARCHIVE TO TAPE ALL DIRECTORIES CREATED ~~ON~~ DURING THIS TASK. THIS WAS THEN SENT TO SITAKANTA.

AN

6/6/95

05/29/95

All files (final and intermediate) created during this numerical exercise have been transferred to two tapes which are a part of this notebook. These two tapes are reference by dates (i.e. 5/22/95 on Tape 1 and 5/29/95 on Tape 2)

WMM 6/6/95

Geochemical constraints for the Akrotiri transport model.

Cu transport from the artifacts was modeled. A groundwater chemistry model was developed to obtain estimates of the solubility of Cu for the source term. Published groundwater chemistry from the semiconfined aquifer immediately beneath the Minoan volcanics at the Karterados site (Kourmoulis, 1990) was used as a basis for the bulk chemistry. Several equilibrium aqueous speciation models were generated at pH 6 to 8 using EQ3 version 7.1 and its associated database DATA0.COM.R16 (Wolery and Daveler, 1992). Equilibrium speciations using the reported HCO^{3-} concentrations resulted in elevated CO_2 pressures that are unrealistic for the near surface. Therefore, the CO_2 pressure was reduced to 10^{-3} bar, which resulted in HCO_3^- concentrations around 10^{-3} molar. The solubility of Cu was assumed to be controlled by atacamite ($Cu_4Cl_2(OH)_6$). Although cuprite and paratacamite have been observed as corrosion products, cuprite is an intermediate product with Cu in the reduced (cupric) state, and no data are available for paratacamite. The suite of aqueous speciation and solubility calculations yielded values for Cu concentrations around 10^{-5} molar, which was used in all simulations as the source term solubility.

Constant equilibrium distribution coefficients (K_d in Table 4) were invoked for all zones. These coefficients are also based on the simple water chemistry model and literature data for Cu sorption on quartz for the tuffaceous units (Schindler et al., 1987) and on kaolinite for the packed earth (Salomons and Förstner, 1984).

References and Table are given in milestone report 06509S-006 of the Natural Analog Project submitted to NRC May 26, 1995.

WMM 6/6/95

I have reviewed this notebook and find it to be generally in compliance with QAP-001. There is adequate information for other qualified people to reproduce the activities.

E. C. Percy

E.C. PEARCY

2/20/97

I am surrendering this Scientific notebook because the project is complete.

Sitakanta Mohanty

2/17/2000

Closed.

E. C. Percy

2/8/2000

Fax 522-5720

1/19

9/20/96

Sensitivity Analyses

Set		Base Case	Min	Max
$\Phi_{s_{1,3}}$	1	0.6	0.6 − 0.18	0.6 + 0.18
		0.52	0.52 − 0.156	.052 + 0.156
Φ_{s_2}	2	0.3	0.30 − 0.09	0.30 + 0.09
$K_{sat_{1,3}}$	1	1.75×10^{-4}	4.02×10^{-6}	7.618×10^{-3}
		1.5×10^{-4}	4.96×10^{-6}	4.536×10^{-3}
K_{sat_2}	2	1×10^{-7}	2.77×10^{-9}	3.6×10^{-6}
$S_{rr_{1,3}}$	1	0.02	0.0	0.04
		0.02	0.0	0.04
	2	0.1	0	0.2
$\alpha_{1,3}$	1	0.5885		
α_2	2	2.7		

Post-it™ Fax Note 7671

Date	# of pages ▶
To Ashok Nedungadi	From S. Mohanty
Co./Dept.	Co.
Phone #	Phone # x5185
Fax # x5720	Fax #

2/19

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1: top & bottom
2: Middle

Sensitivity Analyses

Properties of top & bottom layers are the same and average of the two layers.

Set		Min	Base	Max
1	$\phi_{1,3}$	0.46	0.56	.66
2	ϕ_2	0.20	~~0.15~~ 0.30	~~0.2~~ 0.40
1	K_{sat}	4.02×10^{-6}	1.675×10^{-4} ~~1×10^{-7}~~ m/s	1.0×10^{-3}
2	K_{sat}	1×10^{-8}	1×10^{-7}	1×10^{-6}
1	$S_{wr\,1,3}$	0.0	0.02	0.04
2	$S_{wr\,2}$	0.0	0.1	0.2
1	$\alpha_{1,3}$		0.5885	
2	α_2		2.7	

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9/20/96

5/19

Sensitivity Analyses

Set		Base Case	Min	Max
$\Phi_{s_{1,3}}$	1	0.6 0.52 (1)	(2) 0.6 − 0.18 0.52 − 0.156	(3) 0.6 + 0.18 0.52 + 0.156
Φ_{s_2}	2	0.3	(5) 0.30 − 0.09	(6) 0.30 + 0.09
$K_{cat_{1,3}}$	1	1.75×10^{-4} 1.5×10^{-4}	(8) 4.02×10^{-6} 4.96×10^{-6}	9 7.618×10^{-3} 4.536×10^{-3}
K_{cat_2}	2	1×10^{-7}	(11) 2.77×10^{-9}	(12) 3.6×10^{-6}
$S_{v_{1,3}}$	1	0.02 0.02	(14) 0.0 0.0	(15) 0.04 0.04
	2	0.1	17 0	18 0.2
$\alpha_{1,3}$	1	0.5885		
α_2	2	2.7		

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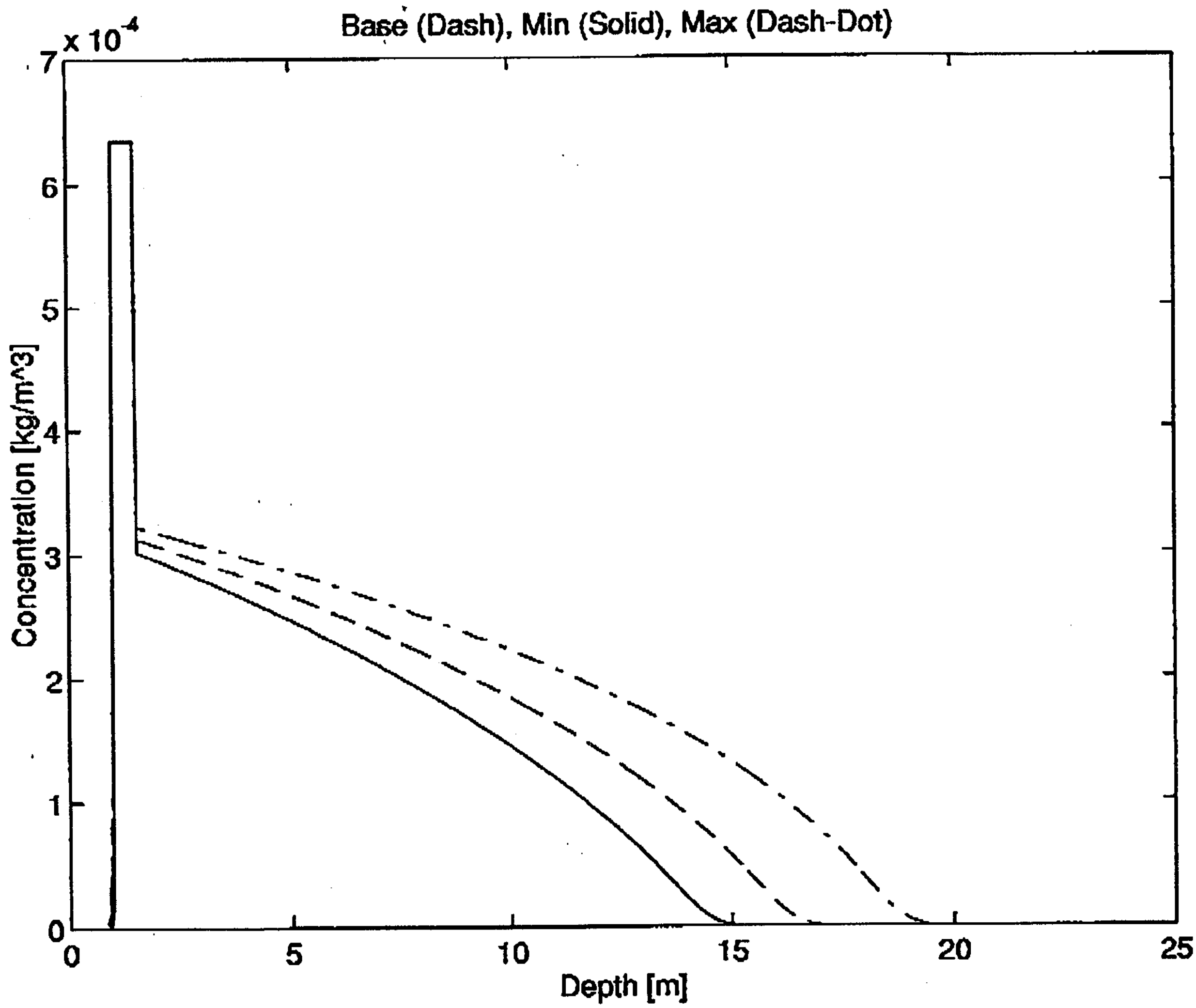
6/19

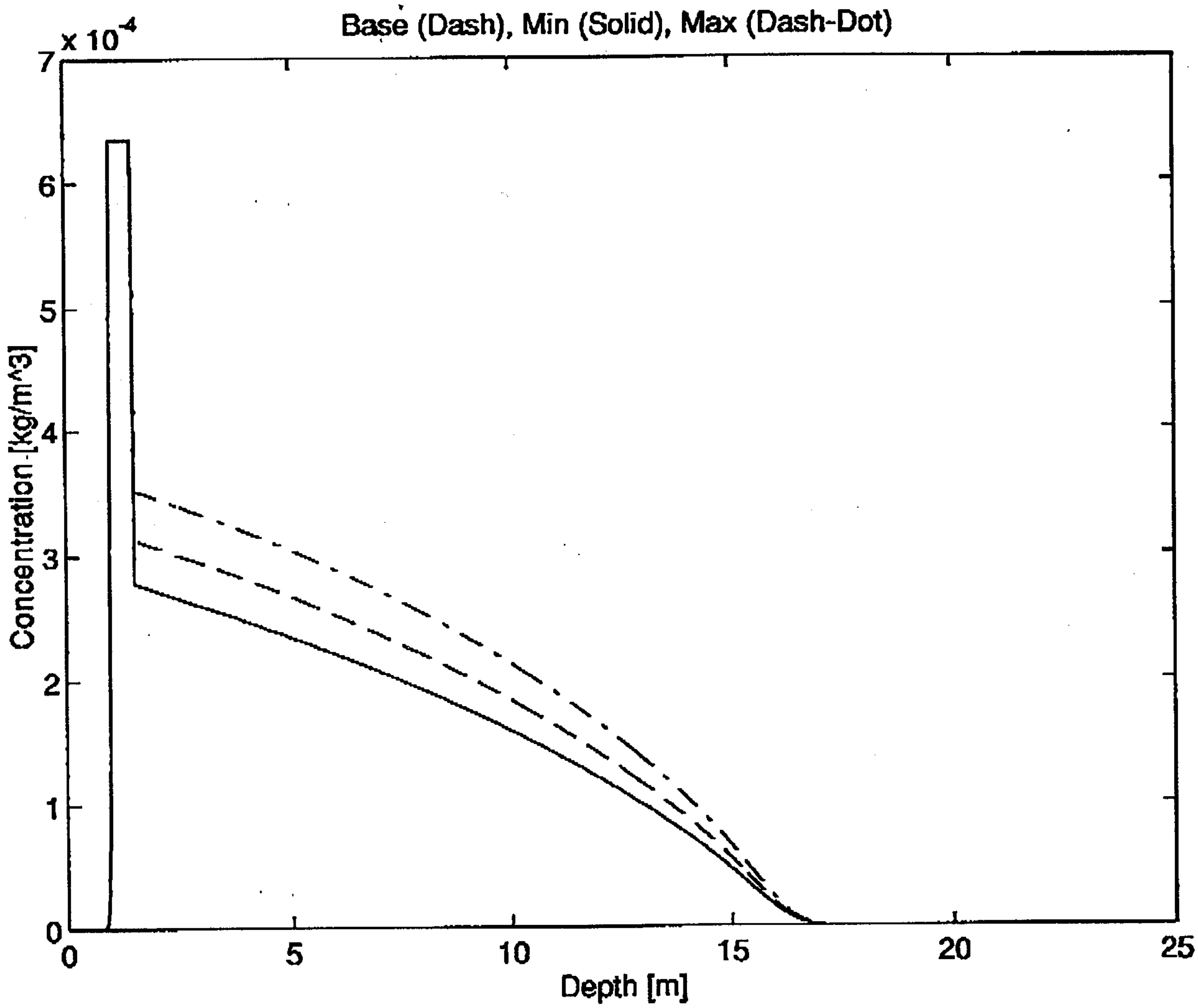
T= 100 years

①, ②, ③



7/19

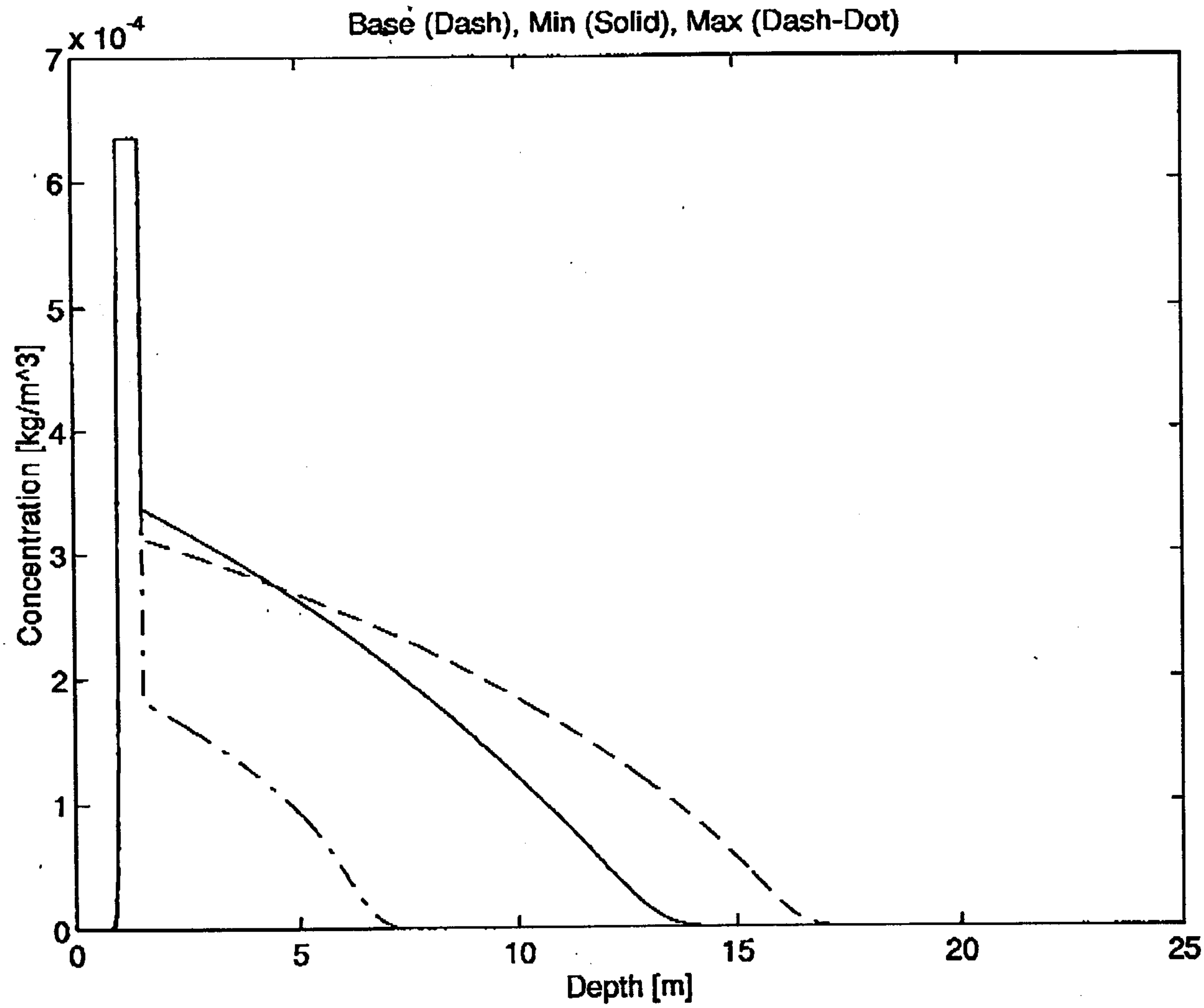
(1), (5), (6)



Base (Dash), Min (Solid), Max (Dash-Dot)
x 10^-4
7
6
5
4
3
2
1
0
Concentration [kg/m^3]
0
5
10
15
20
25
Depth [m]

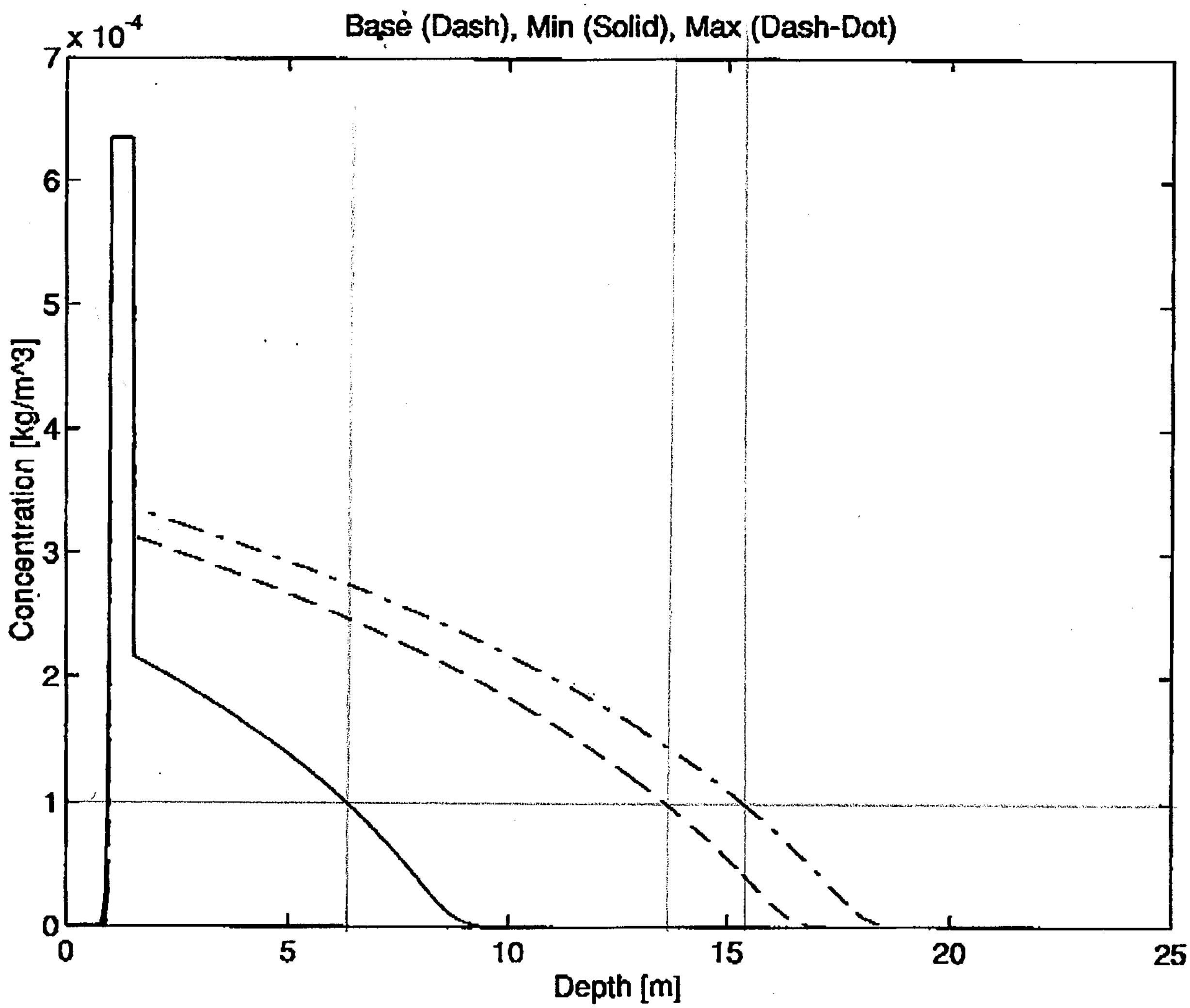
8/19

(7), (8), (9)


Base (Dash), Min (Solid), Max (Dash-Dot)
x 10^-4
7
6
5
4
3
2
1
0
Concentration [kg/m^3]
0
5
10
15
20
25
Depth [m]

9/19

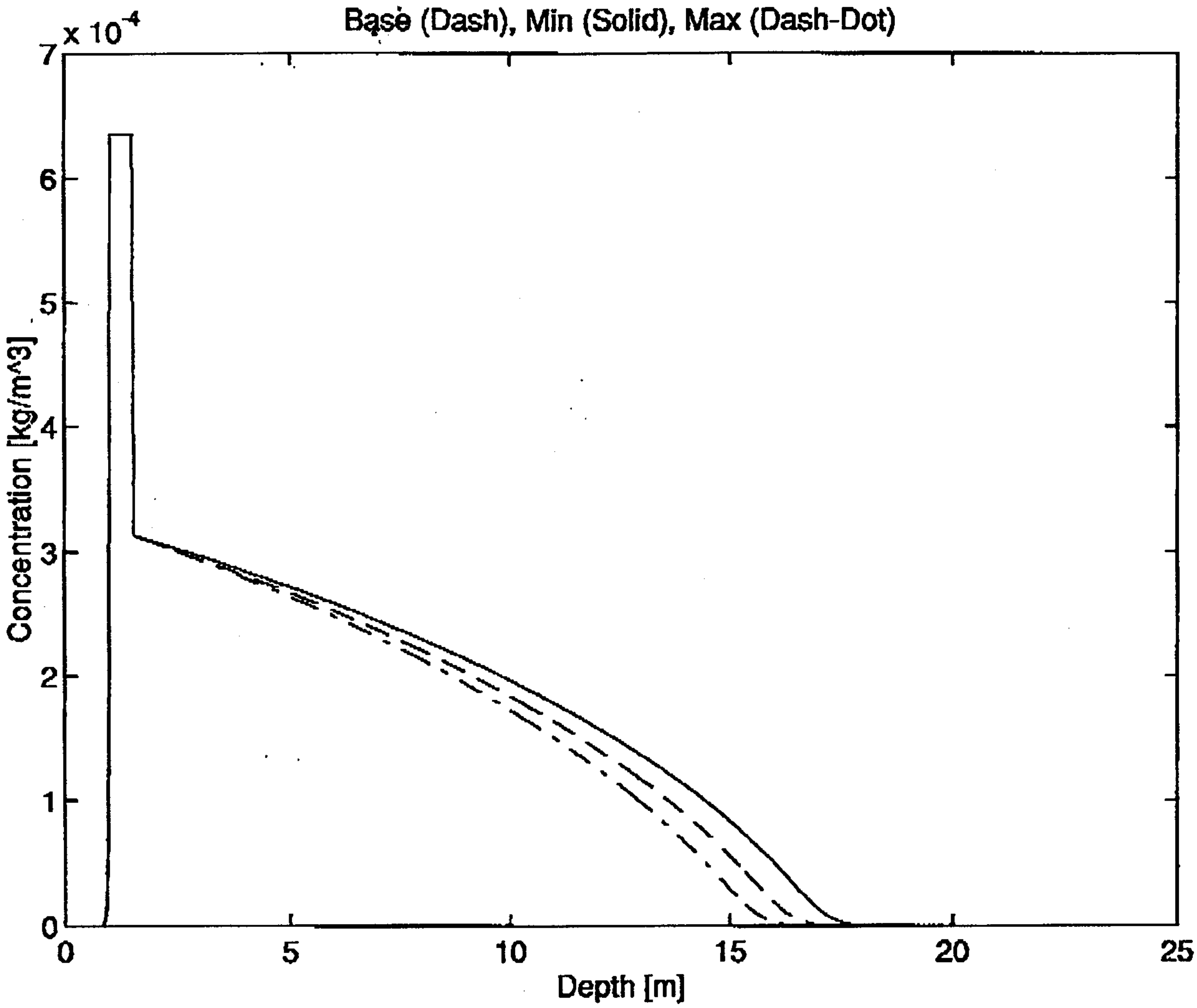
(9), (11), (12)



Min = 6.2 → 54.4 %
base = 13.6
Max = 15.08 → 10.8 %

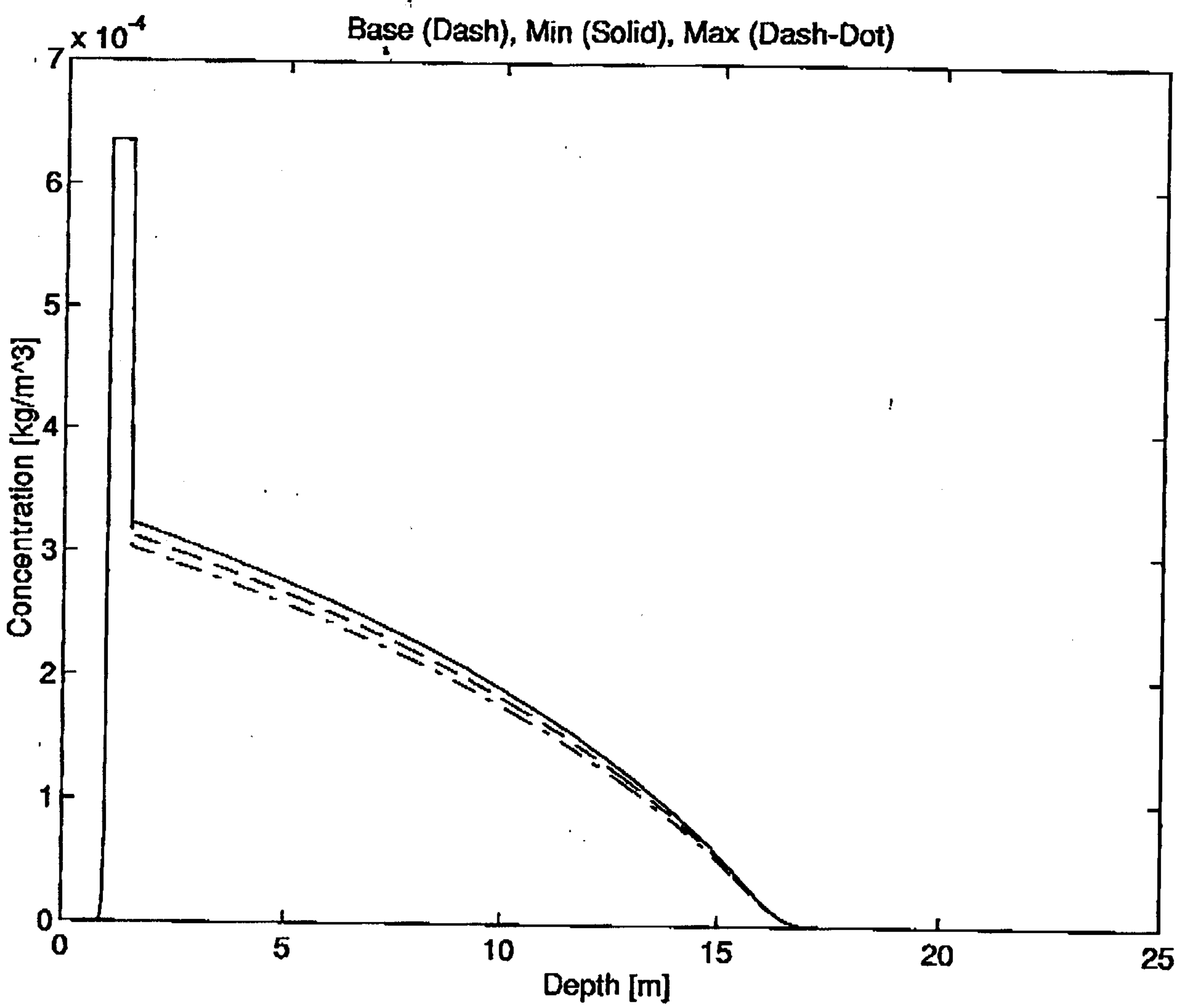
10/19

(1), (14), (15)


Base (Dash), Min (Solid), Max (Dash-Dot)
x 10^-4
Concentration [kg/m^3]
Depth [m]
0
5
10
15
20
25
1
2
3
4
5
6
7

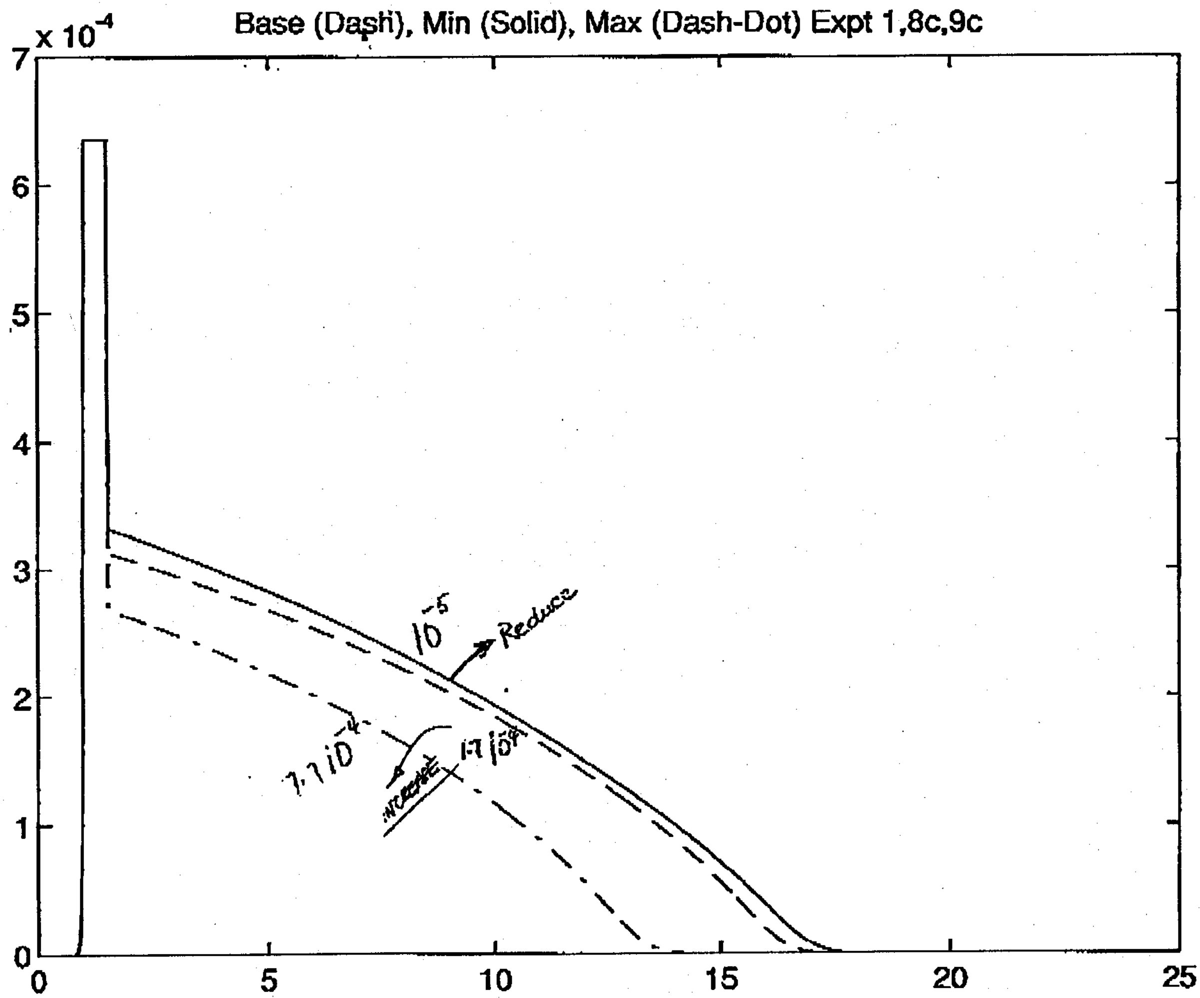
11/19

(5), (17), (18)


Base (Dash), Min (Solid), Max (Dash-Dot)
x 10^-4
7
6
5
4
3
2
1
0
Concentration [kg/m^3]
0
5
10
15
20
25
Depth [m]

12/19

What could be happening ?



DEPTH (m)

MIN	14.0
BASE	13.6
MAX	10.64

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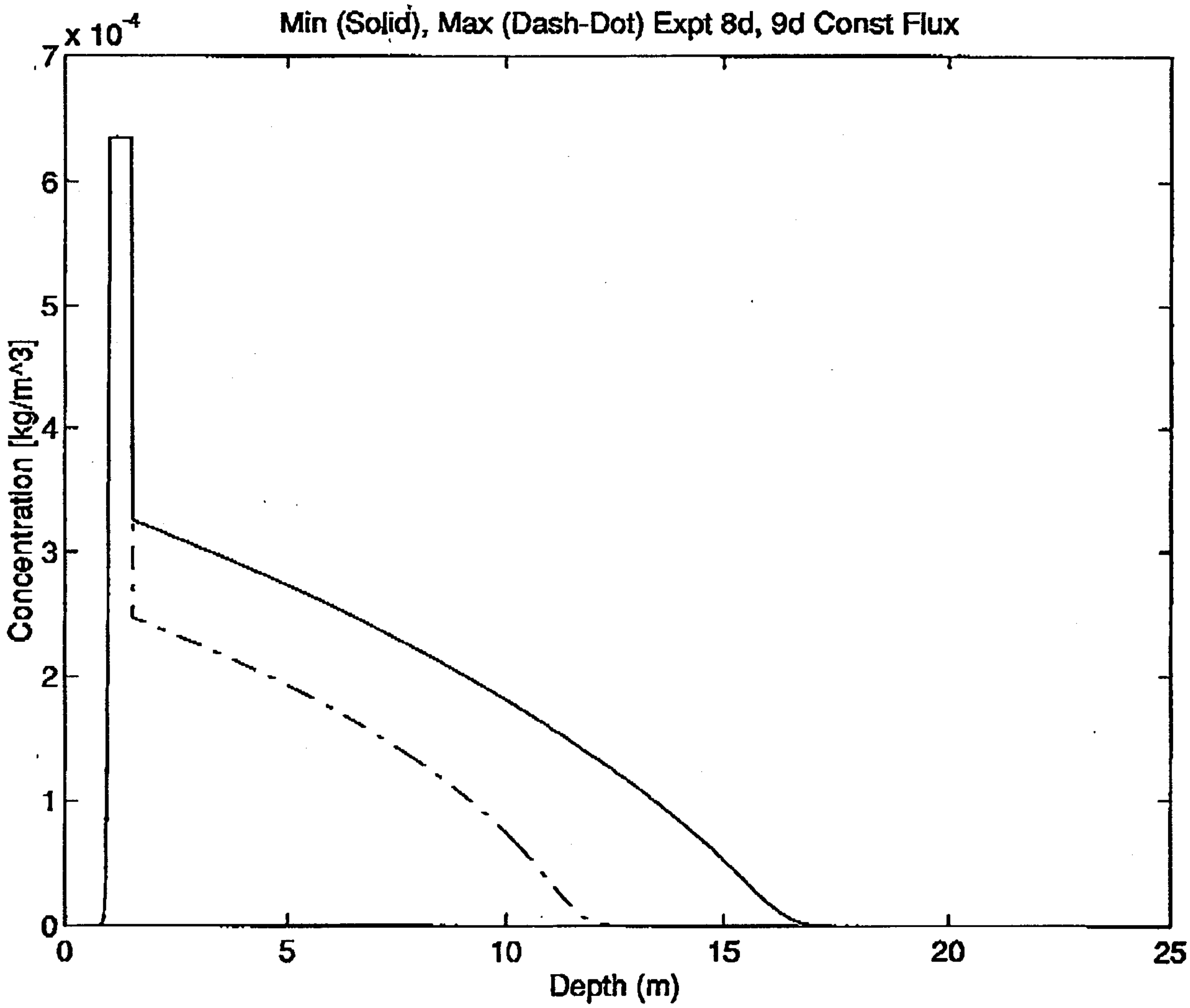
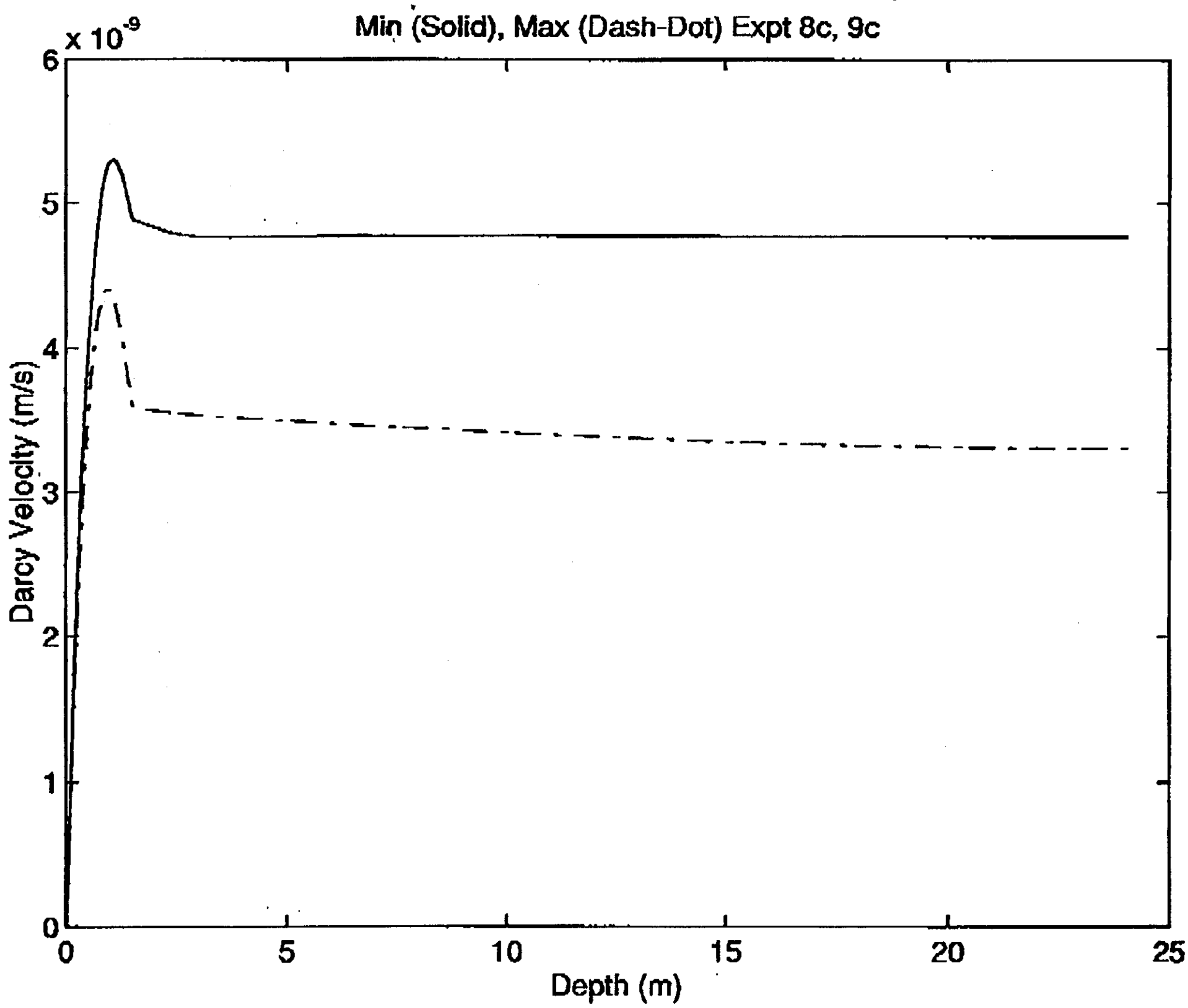
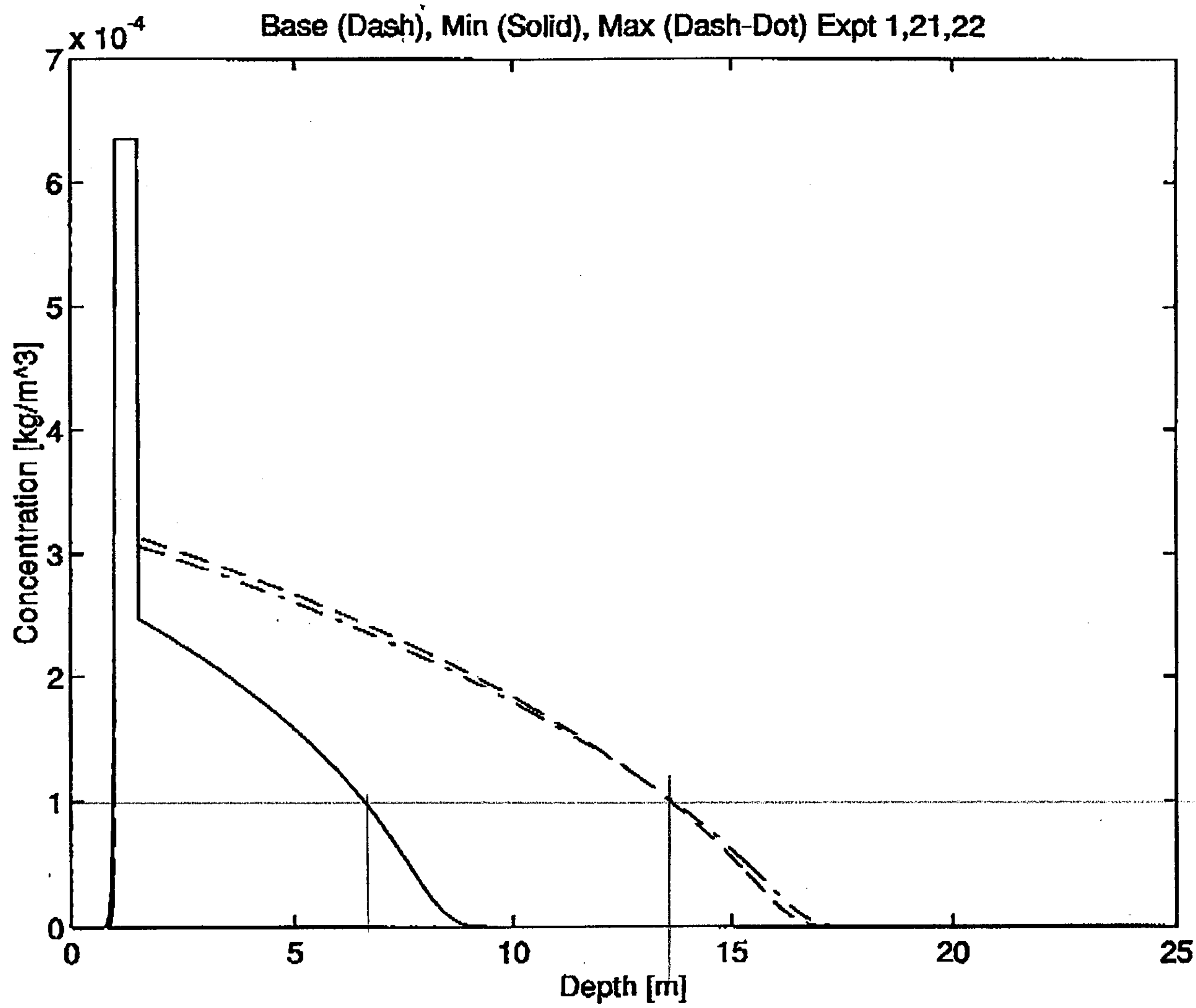

Min (Solid), Max (Dash-Dot) Expt 8d, 9d Const Flux
x 10^-4
7
6
5
4
3
2
1
0
Concentration [kg/m^3]
0
5
10
15
20
25
Depth (m)

15/19

* The smaller Ksat should give
a smaller velocity.
The larger Ksat should give
a larger velocity
But this is not what the code
predicts. Why?



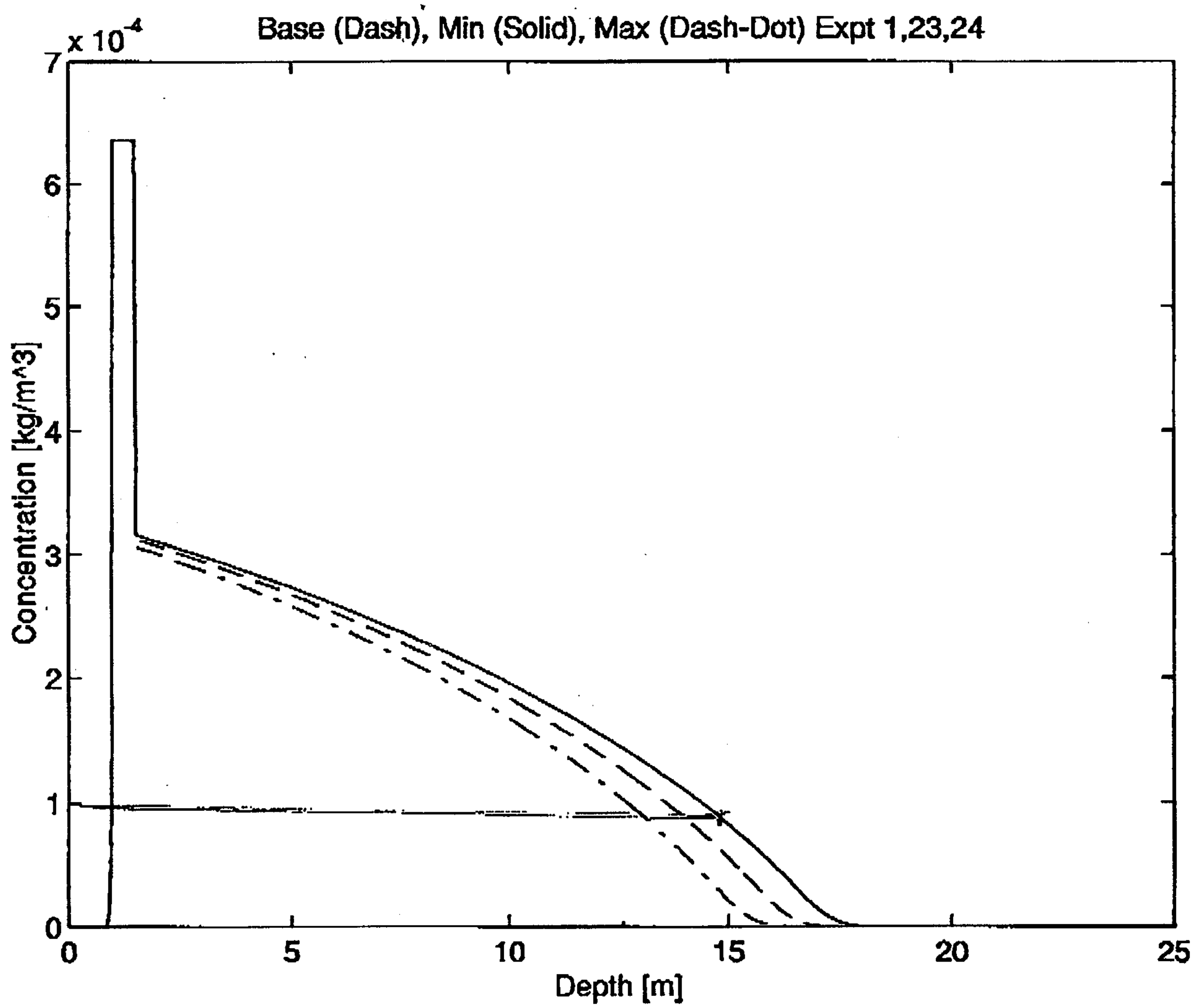
$\alpha_{1,3}$	0.4904	0.207	0.533	16/19
	0.6865	0.4047	0.8193	
α_2	2.7			



	DEPTH (m)	
MIN	6.6	— 51.5%
BASE	13.6	
MAX	13.68	— 0.6%

17/19

	BASE	MIN	MAX
$\alpha_{1,3}$	0.4904 / 0.6865		
α_2	2.7	23 [1.8]	24 [3.6]



	DEPTH (m)
MIN	14.4
BASE	13.6
MAX	12.72

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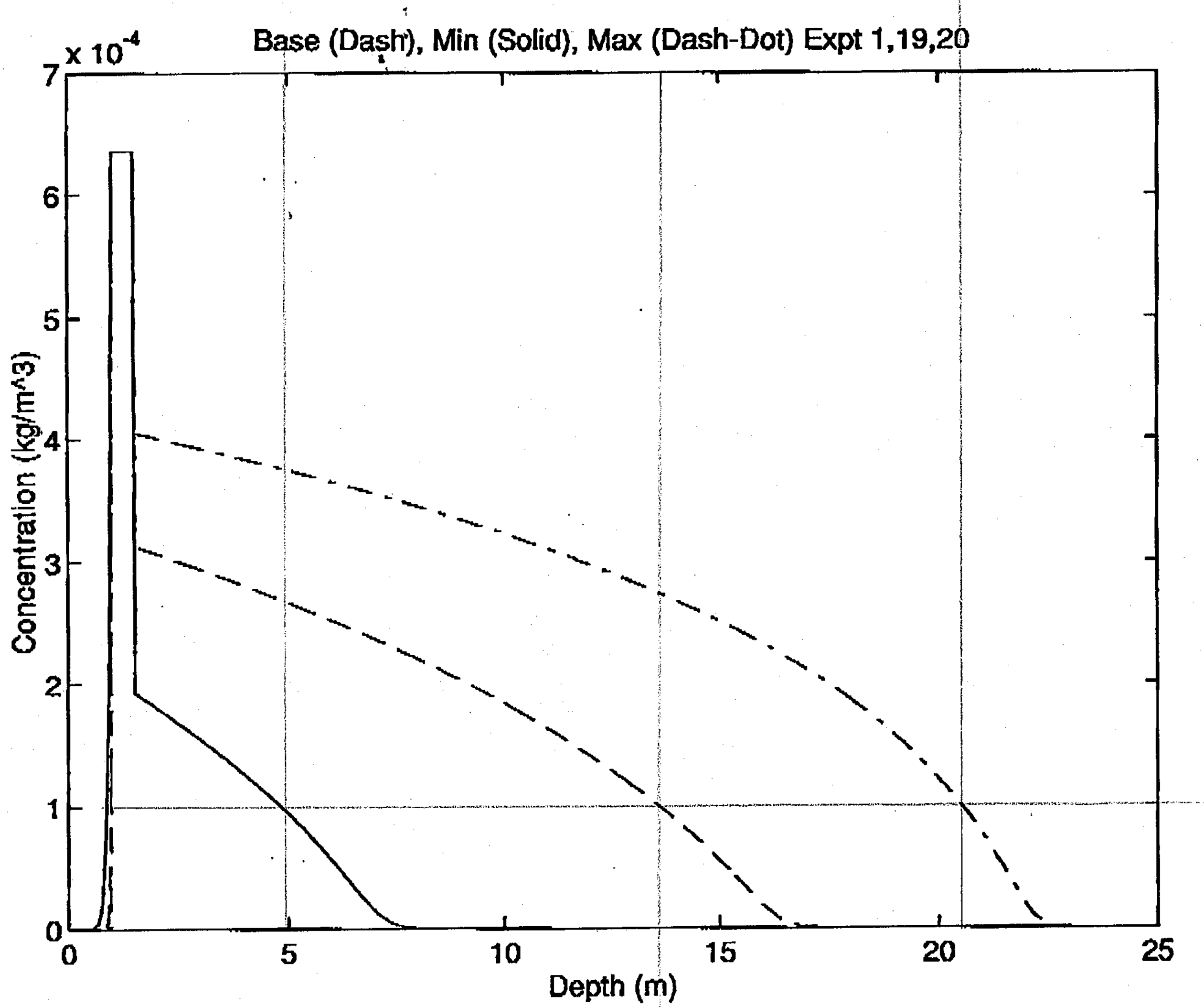

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MAX	20.52	— 51 %

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