

PRELIMINARY RESULTS OF PAH
MEASUREMENTS IN STACK FLUEGAS OF THE
COAL FIRED 4 MWth. TNO-AFBB

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SUMMARY

This report presents the preliminary results of PAH (Polynuclear Aromatic Hydrocarbons) measurements in the stack flue gas of the TNO 4 MWth. coal fired atmospheric fluidised bed boiler.

These PAH measurements were performed during some experiments under standard boiler conditions (bed temperature 850 °C, bed height 1.05 m, fluidisation velocity 1-3 m/s, airfactor 1.2, Ca/S ratio 1-2).

The parameters varied were the type of coal (Polish and U.S. Virginia) and the flyash refiring.

The PAH-total concentrations varied between 96 and 716 ng/nm³. The emission factors for PAH-total varied between 27 and 200 µg/GJ.

The emission factor for Benzo(a)pyrene varied between 0.3 and 3.1 µg/GJ. These emission factors are comparable with those from modern coal fired powerplants.

Because the PAH concentrations measured were of the same order of magnitude as the reference samples no correlation could be made with the parameters varied.

1. INTRODUCTION

The purpose of the research with the TNO 4MWth.AFBB test facility is to demonstrate that industrial plants can be built with good combustion efficiency and environmental pollution as low as possible. Among many research items, the amount of PAH (Polynuclear Aromatic Hydrocarbons) in the flue gas was also investigated. Because only very few investigators have published PAH emission factors of coal fired fluidised bed combustors, TNO has developed its own PAH sampling procedure. With this method TNO has measured the PAH emissions of coal fired small industrial boilers [2] and a pulverised-coal electricity generating boiler [6].

This report presents the preliminary results of PAH measurements in the stack flue gas of the 4 MWth.TNO-AFBB. When the entire research program concerning the relationship between PAH emissions and boiler performance has been completed a full report will be given.

2. DESCRIPTION OF THE TNO 4 MWth TEST FACILITY

The TNO-AFBB is a 4 MWth. coal fired atmospheric fluidised-bed boiler, designed for research on the combustion of coal and other fuels such as lignite, petroleum cokes, oil, and gas.

A flow diagram is given in figure 1.

Limestone and coal are pneumatically transported to the boiler and via nozzles injected underbed into the 2.25 square meter fluidised bed.

Combustion of coal takes place in the 1.05 meter high bed section at 850 °C and in the 3.50 meter high freeboard.

The dust laden fluegas is cooled in the convection passes to about 240 °C and is then cleared of ash in cyclones and a bagfilter.

To improve the combustion efficiency, cyclone ash as well as filter ash is recycled. Thus, combustion efficiencies up to 99.3% are achieved.

Overflow flyash is transported to the flyash removal system.

A more detailed description of the plant can be found in [3] and [4].

3. DESCRIPTION OF THE PAH SAMPLING PROCEDURE

3.1 The sampling train

The sampling train consists of the following components:

- an isolated quartz sampling tube (internal diameter 18 mm).
- a heated teflon coated quartzfibre filter (temperature 170 °C).
- 4 ice-cooled modified Greenburgh-Smith impingers.
- a teflon coated quartzfibre filter (ambient temperature).
- XAD-2 resin between two pieces of polyurethane foam.
- pumpsystem provided with manometers, dry gasmeter, rotameter and temperature indicator.

The whole sampling train (except the pumpsystem) is made of glass. The filters are coated by teflon to avoid possible reactions between PAH and quartzfibre.

3.2 The sampling point

The sampling point is located in the fluegas duct (0.6 m x 0.6 m) between the fabric filter and the induced draught fan (figure 1).

The inlet of the sampling probe is provided with a 90° elbow. This elbow is turned upwards against the direction of the flue gas flow. The sampling is performed under isokinetic conditions, so that gaseous PAH as well as PAH condensed on particulate matter is collected.

3.3 Analytical methods

The teflon coated filters (hot and cold) and the XAD-2 resin are extracted with toluene and the flue gas condensate from the ice-cooled impingers is extracted with cyclohexane.

These extracts are combined and spiked with a known concentration of Dibenzo(a,h)anthracene for quality control.

The concentrations of 21 PAH's are determined with HPLC.

4. RESULTS AND DISCUSSION

The PAH-measurements were carried out during the experiments 79, 83, 140 and 141. The experimental data of these experiments are given in table 1.

Some differences between these experiments, that may have an effect on the PAH concentrations, are:

*type of coal : run 79, 140 and 141 Polish coal 2 and 3
run 83 U.S. coal (Virginia 1)

Coal characteristics are given in table 2.

*refiring flyash : run 79 and 83 no refiring
run 140 and 141 refiring flyash and selective
discharge of filterash.

The flyash refired is a mixture of cyclone ash and fabric filter ash. The maximum flyash refiring flow is about 1500 kg/h and also limited by the capacity of the fabric filter (200 kg/h). Because filter ash has a lower carbon content than cyclone ash, a part of the filter ash flow is discharged to the fly ash containers. During run 141 a combustion efficiency of 99.1% was achieved (table 1).

The PAH sampling was performed according to the time schedule given in table 3. Actual flue gas conditions at the sample point during the sampling periods were: temperature ~200 °C
velocity ~6 m/s

Some boiler characteristics during the sampling periods are given in diagrams (figure 2, 3, 4, 5).

Remarks on the diagrams are:

* figure 2, run 79 : The total coal flow was fed into bed 2.
The combustion air flow was split in a
ratio 40/60.

* figure 3, run 83 : The bed temperature control system was tested.

* figure 5, run 141 : The limestone rotary valve malfunctioned.

Flue gas compositions, molar Ca/S ratio, and airfactor during the sampling periods are given in table 4.

PAH concentrations and emission factors for total PAH and Benzo(a)pyrene are given in tables 5.1 and 5.2.

The emission factors for PAH-total and Benzo(a)pyrene are calculated as follows:

$$\text{factor} = \frac{\text{concentration in ng/nm}^3 * \text{average flue gas flow (dry) in nm}^3/\text{h}}{\text{average gross thermal heat input in GJ/h}}$$

$$\text{average flue gas flow (dry)} = 4000 \text{ nm}^3/\text{h}$$

$$\text{average gross thermal input} = 14.4 \text{ GJ/h}$$

The emission factors for Benzo(a)pyrene are compared in figure 6 with measurements carried out by TNO/ECN [2] with the same sampling procedure. The diagrams in this figure are composed according to data given by Hangebrauck et al. [1].

5. PRELIMINARY CONCLUSIONS

- * The concentrations for PAH-total are 96-716 ng/nm³ (n=11).
The emission factors for PAH total are 27-200 µg/GJ (n=11).
The emission factors for Benzo(a)pyrene are 0.3-3.1 µg/GJ (n=11).
- * The emissions factors for PAH-total are of the same order of magnitude as those given by Veldt et al. [5]. Their average value for PAH-total for coal fired plants is 440 µg/GJ. Their expected value for PAH-total for modern coal fired power plants is about 100 µg/GJ.
- * Due to the fact that the concentrations for PAH-total in the stack flue gas samples and the reference samples are of the same order of magnitude, no correlations can be made between the PAH emissions and experimental parameters as type of coal, airfactor, CO and C_xH_y concentrations and the influence of fly ash recycling.

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TABLE 1. TNO 4MW ATMOSPHERIC FLUIDISED BED BOILER TEST FACILITY

Summary of experimental data

RUN NUMBER			079	083	140	141	
START: date			yyymmdd	840111	840125	850221	850313
time			hr:min	11:45	5:00	16:00	20:00
END : date			yyymmdd	840112	840126	850222	850314
time			hr:min	09:00	08:30	09:00	08:00
TYPE OF EXPERIMENT			d,s,t	s	s	s	s
FLUID VELOCITY			m/s	2.2	2.1	2.19	2.40
AIR FACTOR				1.25	1.13	1.22	1.23
BED HEIGHT			cm	107	106	106	107
BED SECTIONS FLUIDISED			1 or 2	2	2	2	2
CYCLONES			1,M,s	1,M,s	M	M,s	M,s
TYPE OF COAL				polish2	va,usa	polish3	polish3
COAL FLOW	bed 1	kg/hr	0	247	219	236	
	under/over			u	u	u	
	bed 2	kg/hr	481	231	223	239	
	under/over		u	u	u	u	
GAS FLOW	bed 1	m^3/hr	-	-	-	-	
	bed 2	m^3/hr	-	-	-	-	
	air preheaters	m^3/hr	-	-	-	-	
TYPE OF SORBENT *				1	1	1	1
SORBENT FLOW	bed 1	kg/hr	0	23	0	0	
	bed 2	kg/hr	14	26	22	12	
Ca/S RATIO (molar)				1.5	2.2	2.4	1.2
SULFUR RETENTION			%	37	77	85	54
REFIRING cyclones			+/-	-	-	+	+
fabric filter			+/-	-	-	+ 1)	+ 1)
CYCLONE DUST FLOW			kg/hr	41	68	970	1300
Unburned carbon			%	15.4	42.7	12.8	13.0
FABRIC FILTER DUST FLOW			kg/hr	59	50	110	200
Unburned carbon			%	25.4	25.8	14.3	12.5
FLY ASH COLLECTED			kg/hr	100	118	58	50
Unburned carbon			%	n.m.	n.m.	11.7	10.3
BED ASH COLLECTED			kg/hr			4	2
COMBUSTION EFFICIENCY			%	n.m.	n.m.	98.7	99.1
combustion in bedsection			%	n.m.	n.m.	91.6	91.5
combustion in freeboard			%	n.m.	n.m.	7.1	7.6
energyloss in flyash			%	n.m.	n.m.	1.3	0.9
TEMPERATURES			deg C				
air preheater 1				60	60	59	59
air preheater 2				66	63	60	60
bed 1				857	843	842	843
bed 2				883	850	849	849
mid freeboard				884	846	869	871
end freeboard						902	911
flue gas conv. section				355	357	424	440
flue gas outlet boiler				227	227	246	253
flue gas before filter				213	213	225	232
STEAM							
steam pressure			barg	14.0	14.0	14.0	14.0
steam flow main drum			ton/hr	2.55	2.43	3.27	3.64
steam flow side drum			ton/hr	2.48	2.47	2.39	2.40
LOAD (fuel input)			MW	3.89	4.06	3.82	4.10
POWER (steam output)			MW	2.55	2.43	3.06	3.26
Heat Exchanger							
test tubebank AKZO version				2	2	4	4
HE side drum				bed1&2	bed1&2	bed1&2	bed1&2
Heattransfer coeff. bed -> tubebank			W/m^2K	n.m.	n.m.	286	285
STACK EMISSIONS							
O2 (Ametek)			%	3.7	4.0	5.2	4.8
O2 (GAR)			%	3.8	4.3	4.2	4.4
SO2			ppm,g/GJ	310 267 288	236 168	62 261	241
NOx			ppm,g/GJ	328 192 262	146 235	156 211	140
CxHy			ppm,g/GJ	37 91	7 21	10 31	10 41
CO			ppm,g/GJ	362 130 214	72 292	110 303	114
CO2			%	14.6	14.5	14.4	14.0

*) 1 = Engis 1,5-2,5 mm

n.m. = not measured

1) Selective discharge of filter ash

TABLE II

Characterisation coals and limestones

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Coal type		Polish 1	Polish 2	Americ 1 (Virg)	Polish 3	Polish 4
Moisture	(%)*	3.1	2.5	3.1	3.0	1.5
Ash	(on dry)(%)	15.1	15.6	9.8	9.5	9.0
Volatile	" (%)	25.3	29.2	28.1	31.9	31.4
C	" (%)	69.0	70.3	75.9	76.4	75.9
O	" (%)	10.3	7.5	6.6	7.0	8.3
H	" (%)	4.2	4.6	4.4	5.0	4.4
N	" (%)	1.2	1.2	1.5	1.4	1.7
S	" (%)	0.85	0.66	1.61	0.76	1.06
Cl	" (%)	-	-	-	-	-
HHV @	" (MJ/kg)	28.45	29.01	30.65	31.72	30.90
Volatile (DAF)\$	(%)	29.8	34.6	31.2	35.3	34.5
C	" (%)	81.2	83.3	84.2	84.4	83.4
O	" (%)	12.1	8.9	7.3	7.7	9.1
H	" (%)	4.9	5.5	4.9	5.5	5.3
N	" (%)	1.4	1.4	1.7	1.5	1.9
S	" (%)	1.0	0.78	1.79	0.84	1.16
Cl	" (%)	-	-	-	-	-
Particle size distribution						
< 10 mm	(%)	100.0	99.9	97.5	99.1	99.3
< 8 mm	(%)	100.0	99.7	90.2	95.1	96.1
< 6.3 mm	(%)	99.9	99.3	81.4	85.2	86.1
< 5.6 mm	(%)	98.5	97.4	75.8	80.9	82.3
< 4 mm	(%)	87.9	84.1	63.6	68.2	69.2
< 2.8 mm	(%)	65.1	67.7	53.7	56.5	59.8
< 2 mm	(%)	53.1	54.5	45.1	40.4	47.8
< 1 mm	(%)	35.3	34.4	29.4	24.4	32.1
< 0.5 mm	(%)	27.5	23.4	18.8	11.5	22.0
< 0.2 mm	(%)	18.1	13.6	9.1	5.3	14.9

* percentages on weight base

\$ DAF - dry and ash free

@ HHV - gross heating value

Table 3. Time schedule for PAH sampling 4 MWth. TNO-AFBB

RUN NR	Sample	Sample code	date	time	nm3
079	I	41500079	840111	10.06-12.25	7.77
	II	41501079	840111	12.43-14.53	7.27
	III	41502079	840111	15.21-17.58	8.70
083	I	41503083	840125	10.17-12.32	3.46
	II	41504083	840125	12.57-15.52	8.52
	III	41505083	840125	16.09-18.53	8.78
140	I	41508140	850221	11.10-14.10	6.26
	II	41509140	850221	14.40-17.40	6.47
	III	41510140	850222	09.05-12.10	6.53
141	I	41511141	850313	10.30-13.30	7.80
	II	41512141	850314	10.35-14.00	4.71
	III				

Table 4. Fluegas composition, molar Ca/S ratio and airfactor during PAH measurements in stack fluegas 4 MWth. TNO-AFBB.

RUN NR	79			83			140			141	
SAMPLE	I	II	III	I	II	III	I	II	III	I	II
SAMPLE	41	41	41	41	41	41	41	41	41	41	41
CODE	500	501	502	503	504	505	508	509	510	511	512
	079	079	079	083	083	083	140	140	140	141	141
SO ₂	247	204	294	281	298	306	68*	70	76	261*	261*
NO _x	313	358	325	306	277	268	235*	240	231	211*	211*
C _x H _y	15	16	31	8	7	7	10*	9	7	10*	10*
CO	259	261	341	220	214	210	292*	296	294	303*	303*
O ₂	4.2	4.6	4.0	4.4	4.2	4.3	4.2*	4.1	4.4	4.4*	4.4*
CO ₂	14.4	14.1	14.4	14.5	14.6	14.6	14.4*	14.5	14.5	14.0*	14.0*
Ca/S	1.8	1.6	1.4	2.1	2.2	2.1	2.4*	2.4	2.4	1.2*	1.2*
Airfact.	1.25	1.29	1.22	1.14	1.13	1.12	1.22*	1.23	1.22	1.23*	1.23*

Remarks: Concentrations of SO₂, NO_x, C_xH_y and CO in cm³/nm³ (vppm)

Concentrations of O₂ and CO₂ in %

*During this PAH sampling periode the fluegas composition was not measured at the boiler outlet but in the freeboard. Therefore the average values for the whole run are given.

Table 5.1

PAH measurements in stack flue gas 4 MW AFBB TNO

(concentrations in ng/nm³)

		Polish coal 2			US coal (Virg.1)		
		RUN 079			RUN 083		
PAH		I	II	III	I	II	III
1	Phenantrene	110	74	150	203	64	47
2	Anthracene	32	17	24	9	13	6
3	Fluoranthene	20	35	55	46	17	10
4	Pyrene	[2]*	12	16	33	14	6
5	3,6-dimethylphenantrene	[1]	[1]	[1]	[2]	[1]	[1]
6	Triphenylene	[2]	[2]	[3]	[6]	2	[2]
7	Benzo(b)fluorene	[1]	[1]	[2]	[5]	2	[2]
8	Benzo(a)anthracene	[1]	[1]	14	[1]	3	2
9	Chrysene	[1]		5	[4]	2	[1]
10	Benzo(e)pyrene	[4]	11	[3]	[7]	3	[3]
11	Benzo(j)fluoranthene	[16]	[17]	[14]	28	5	[4]
12	Perylene	[1]	3	[1]	[1]	[1]	[1]
13	Benzo(b)fluoranthene	4	11	10	3	2	2
14	Benzo(k)fluoranthene	3	6	5	1	1	1
---> 15	Benzo(a)pyrene	2	11	1	3	2	1
16	Dibenzo(a,j)anthracene	2	[2]	[2]	8	1	2
17	Dibenzo(a,i)pyrene	[1]	[1]	[1]	[2]	2	[1]
18	Benzo(g,h,i)perylene	[3]	[3]	[2]	[4]	2	5
19	Dibenzo(a,h)anthracene						
20	Indeno(1,2,3-cd)pyrene	[2]	14	[1]	[2]	1	[1]
21	3-Methyl-cholantrene	[1]	[1]	[1]	[1]	[1]	[1]
22	Anthanthrene	[1]	3	[1]	[1]	[1]	[1]
PAH concentration in ng/nm ³		206	224	310	367	134	96
PAH emission in µg/GJ		58	63	87	103	37	27
BaP emission in µg/GJ		0.6	3.1	0.3	0.9	0.4	0.3

Remarks: PAH nr.19 is used as an internal standard.

*The values between brackets are the original concentrations of the specific PAH in the stack fluegas sample.

These values are not corrected because the concentration of the corresponding PAH in the reference sample is of the same or even higher order of magnitude.

Table 5.2.

PAH measurements in stack flue gas 4 MW AFBB TNO

(concentrations in ng/nm³)

		Polish coal 3			Polish coal 3		
		RUN 140			RUN 141		
PAH		I	II	III	I	II	III
1	Phenanthrene	[366]*	[211]	[214]	[177]	445	
2	Anthracene	[1]	10	[3]	3	8	
3	Fluoranthene	19	[16]	36	6	10	
4	Pyrene	4	27	2	[29]	46	
5	3,6-dimethylphenanthrene	2		[4]	[4]	8	
6	Triphenylene			22			
7	Benzo(b)fluorene	8	1	[7]	[12]	34	
8	Benzo(a)anthracene	[3]	[3]	[1]	[4]	1	
9	Chrysene	[6]	[9]	8	[3]	3	
10	Benzo(e)pyrene	[13]	73	[6]	[3]	7	
11	Benzo(j)fluoranthene	8	[3]		[8]	[38]	
12	Perylene		[1]	[1]	[1]		
13	Benzo(b)fluoranthene	[2]	[2]	6	[2]	5	
14	Benzo(k)fluoranthene	[1]	[1]	3	[1]	2	
---> 15	Benzo(a)pyrene	[2]	[1]	[3]	[1]	[3]	
16	Dibenzo(a,j)anthracene	[2]	7	16	[2]	[1]	
17	Dibenzo(a,i)pyrene	[24]	[15]	[3]	[5]	[14]	
18	Benzo(g,h,i)perylene	29	[74]	[66]	[56]	[76]	
19	Dibenzo(a,h)anthracene						
20	Indeno(1,2,3-cd)pyrene					[15]	
21	3-Methyl-cholantrene		1		[1]		
22	Anthanthrene						
PAH concentration in ng/nm ³		490	455	401	318	716	
PAH emission in µg/GJ		136	127	112	88	200	
BaP emission in µg/GJ		0.4	0.3	0.7	0.3	0.7	

Remarks: PAH nr.19 is used as an internal standard.

*The values between brackets are the original concentrations of the specific PAH in the stack fluegas sample.

These values are not corrected because the concentration of the corresponding PAH in the reference sample is of the same or even higher order of magnitude.

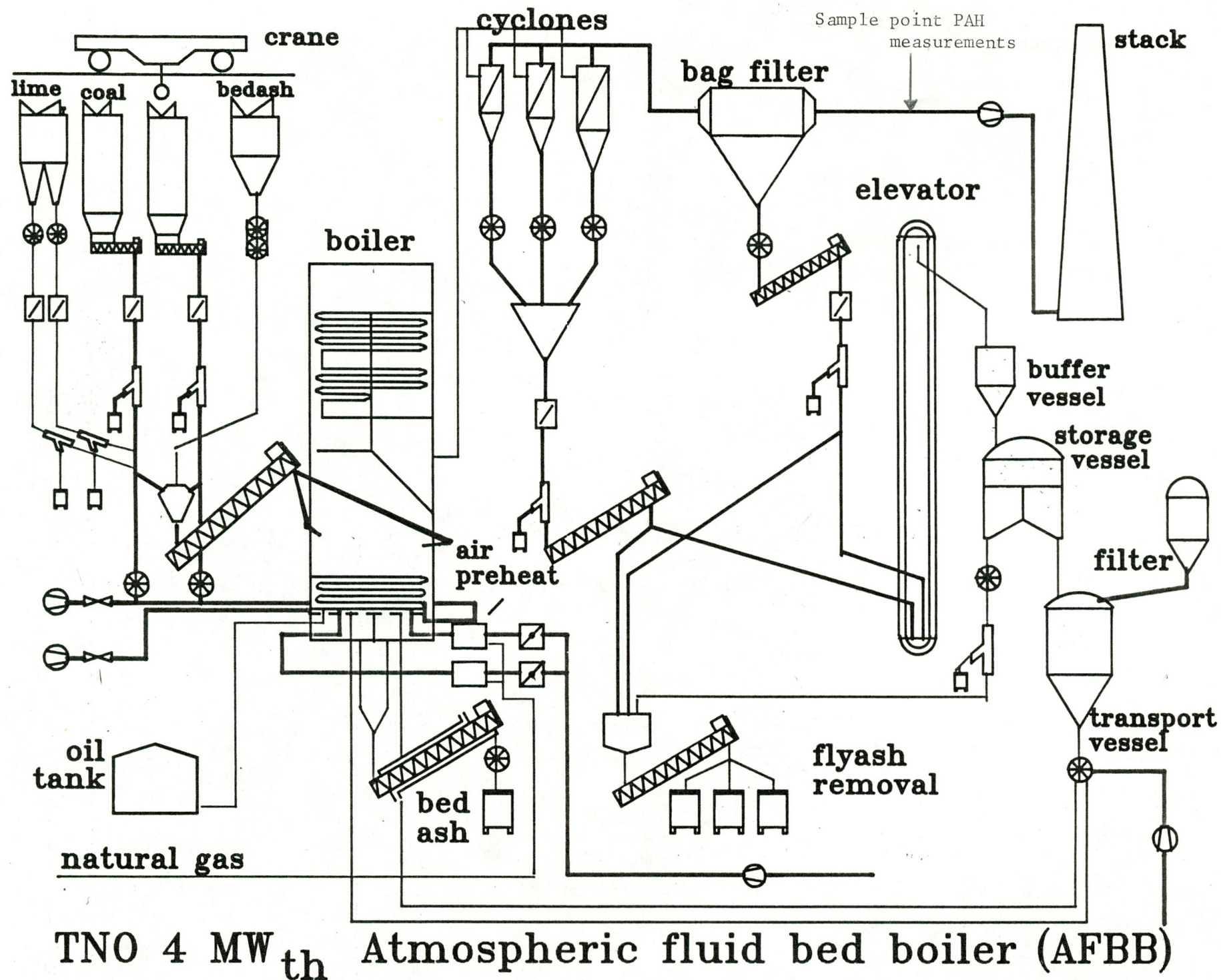
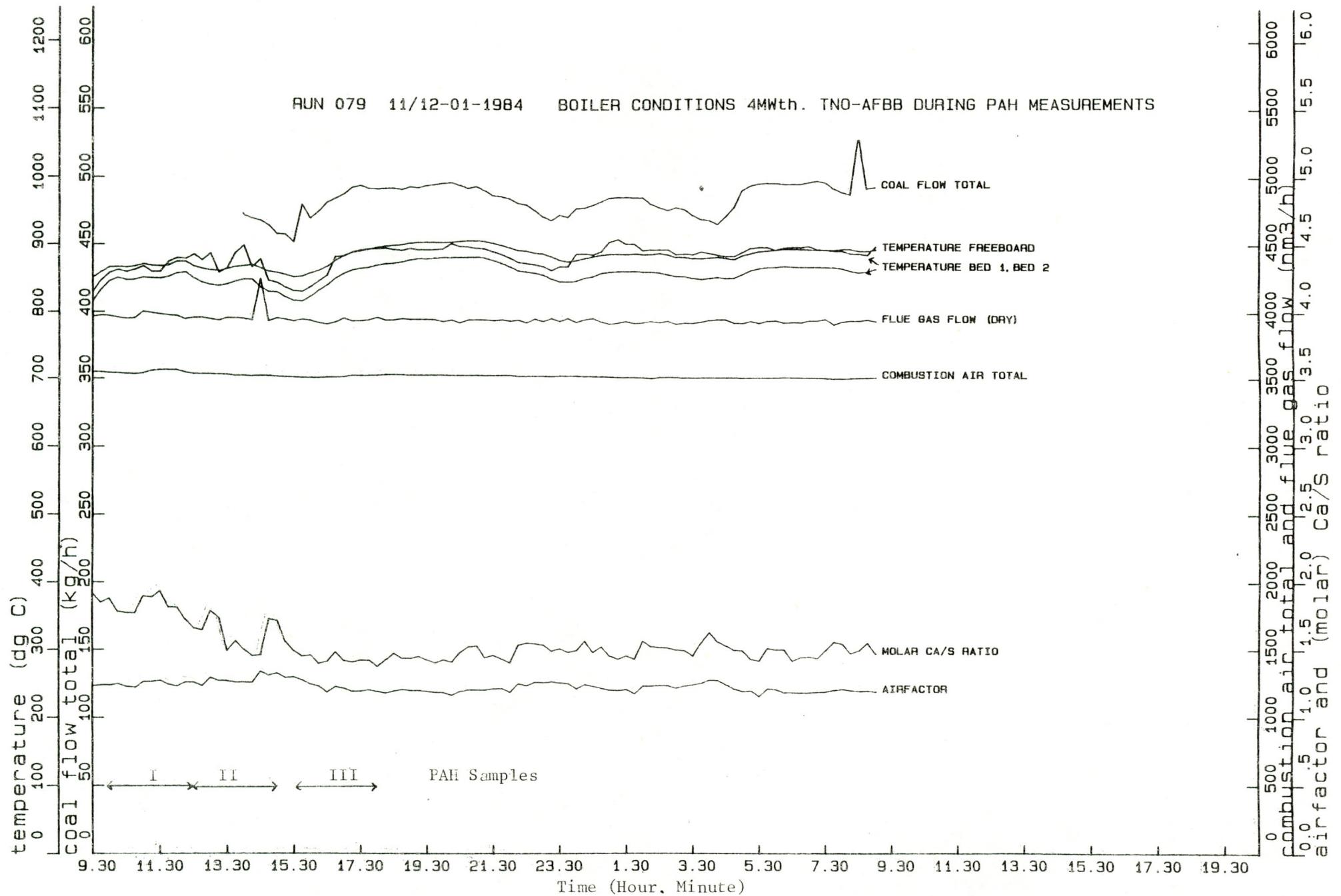


FIGURE II



(continued)

[illegible]

FIGURE III

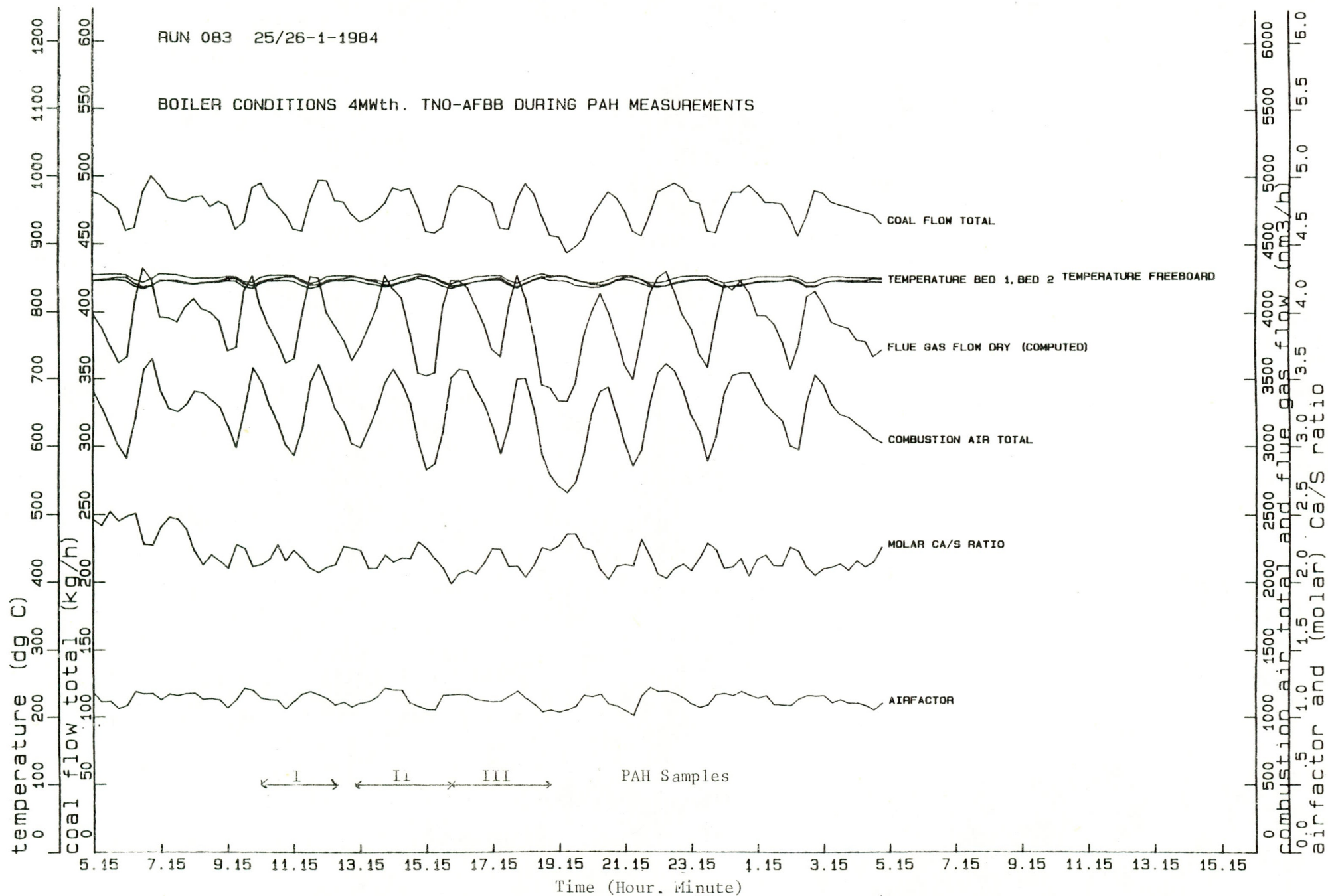
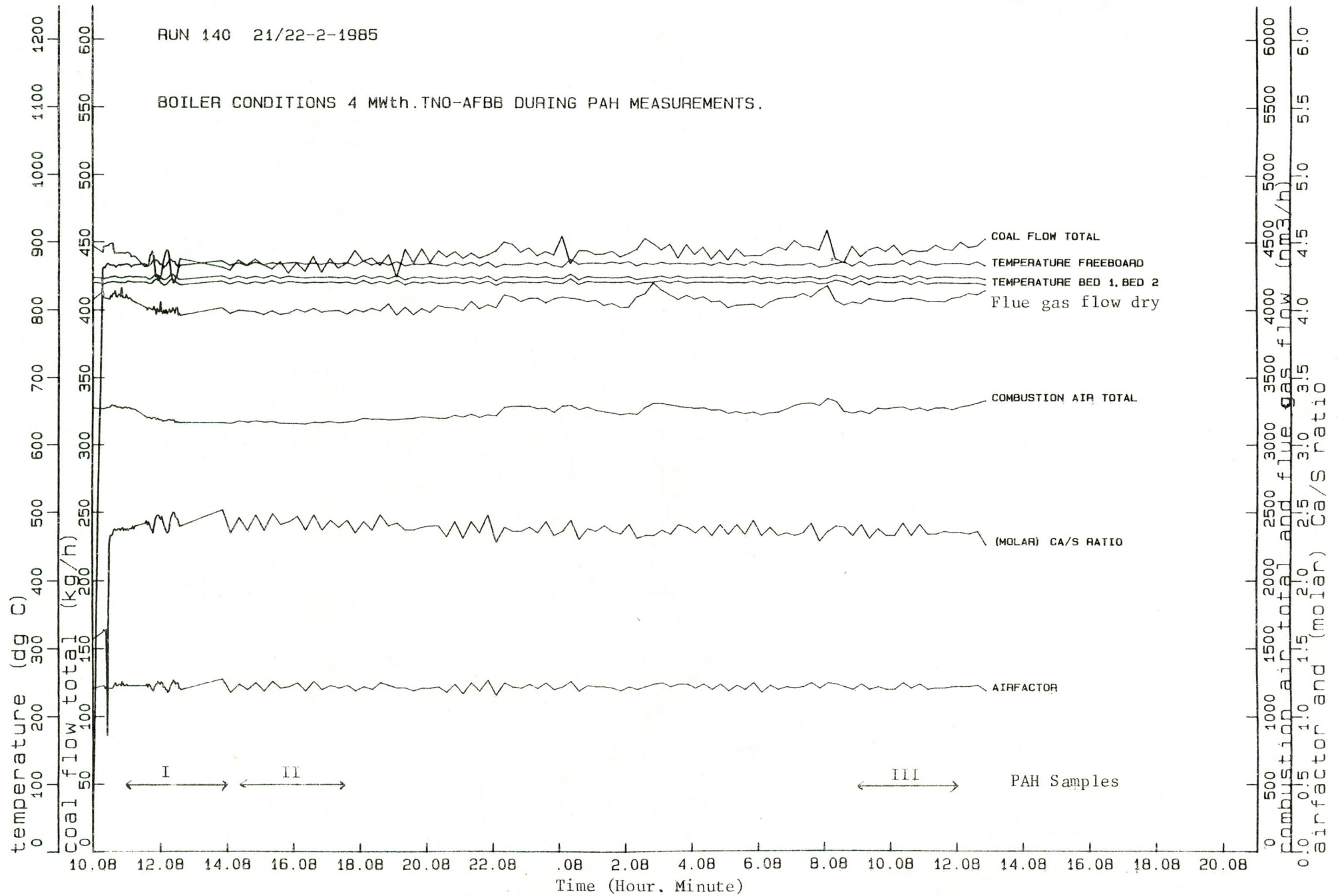


FIGURE IV

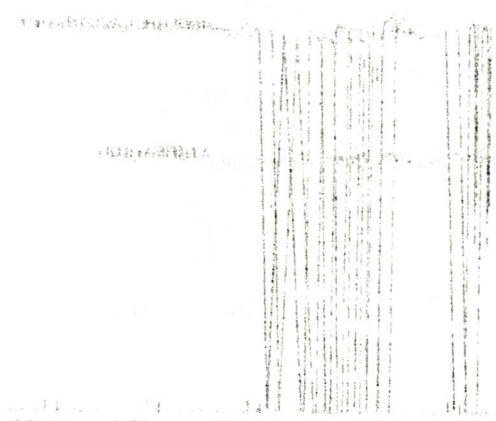


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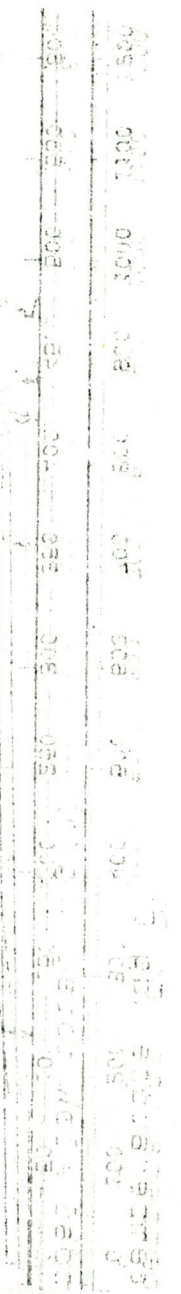
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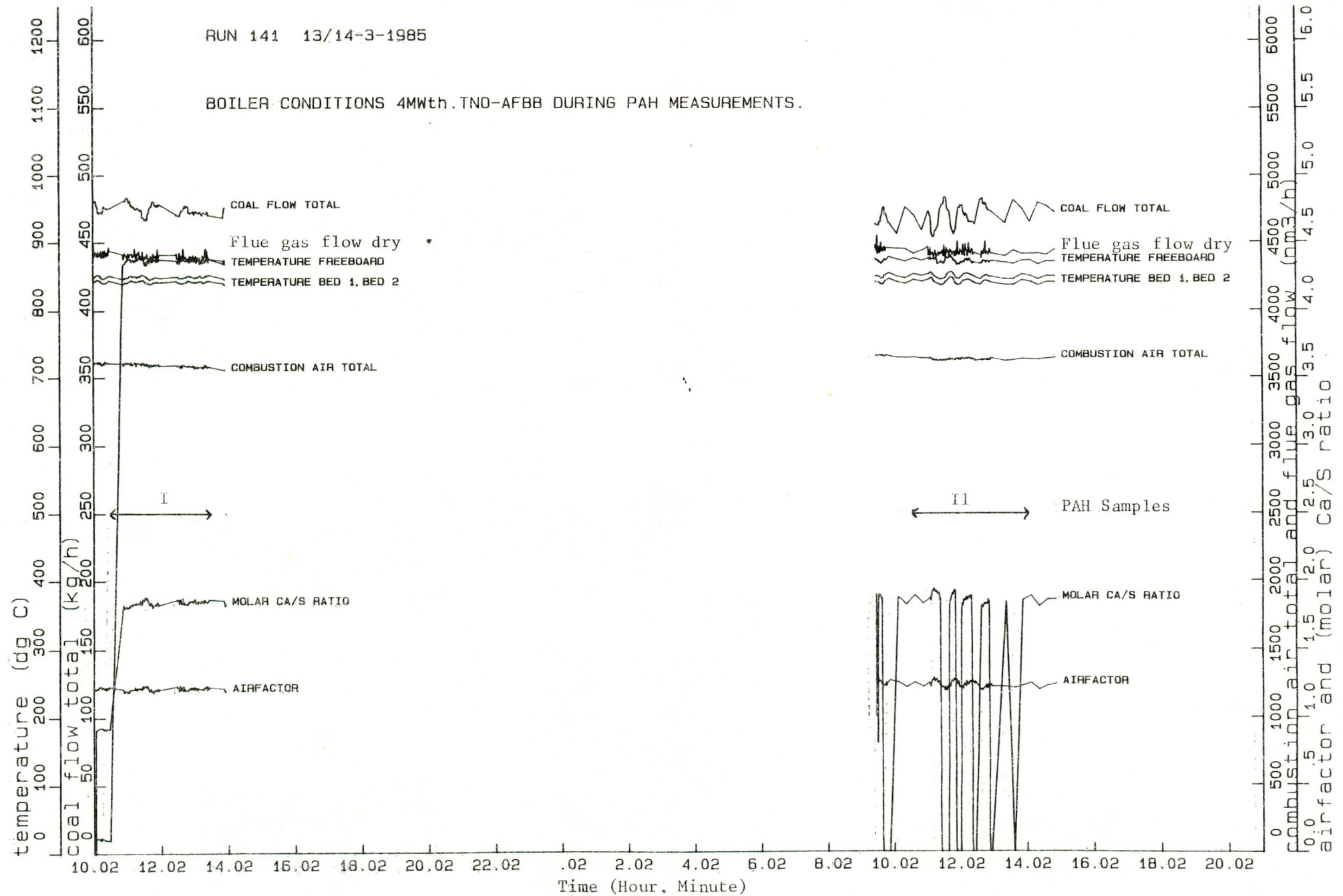
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FIGURE V



1. The first part of the paper is devoted to a discussion of the

mathematical model of the system.

2. The second part of the paper is devoted to a discussion of the

experimental results and their interpretation.

3. The third part of the paper is devoted to a discussion of the



4. The fourth part of the paper is devoted to a discussion of the

conclusions.

FIGURE VI

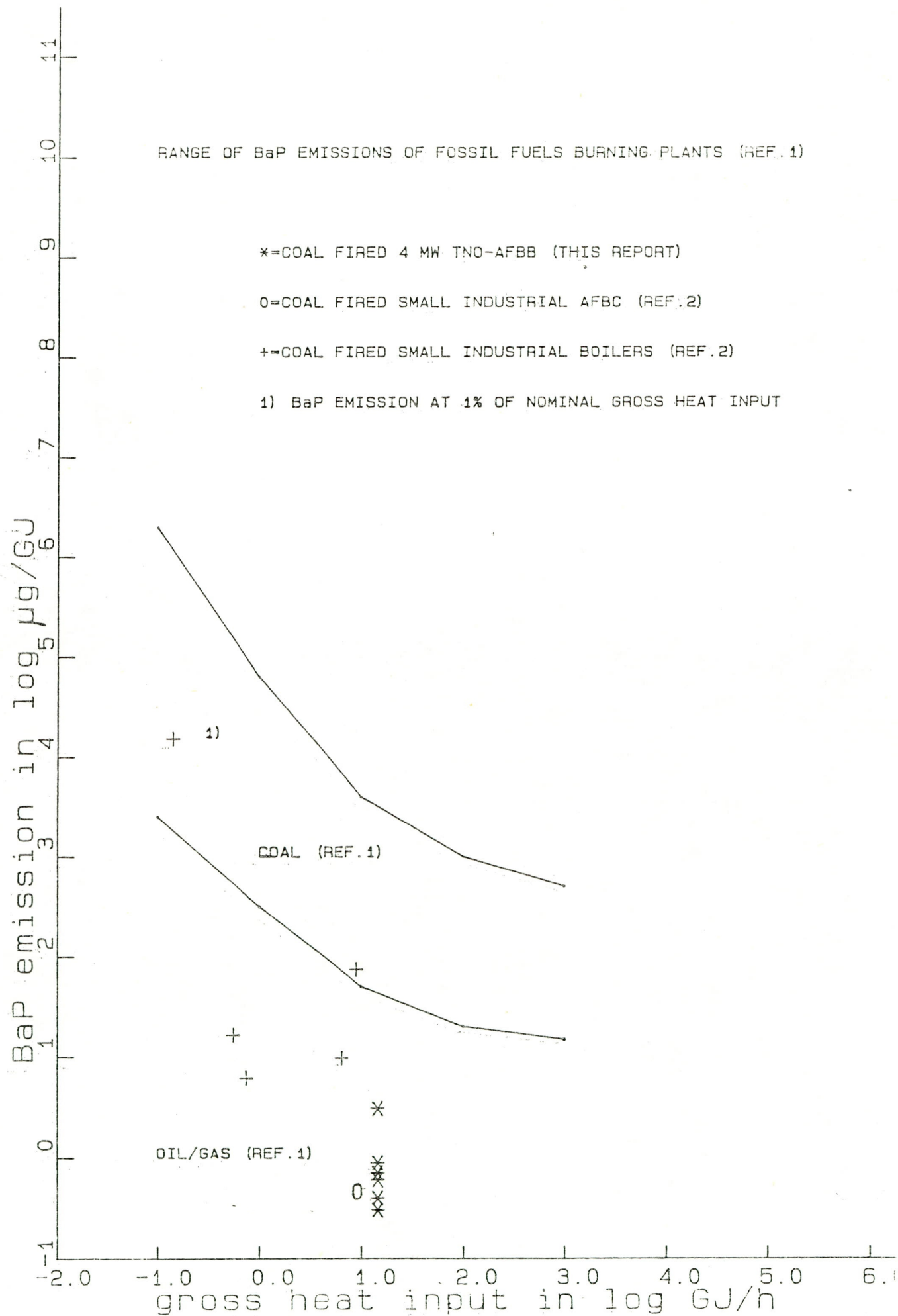
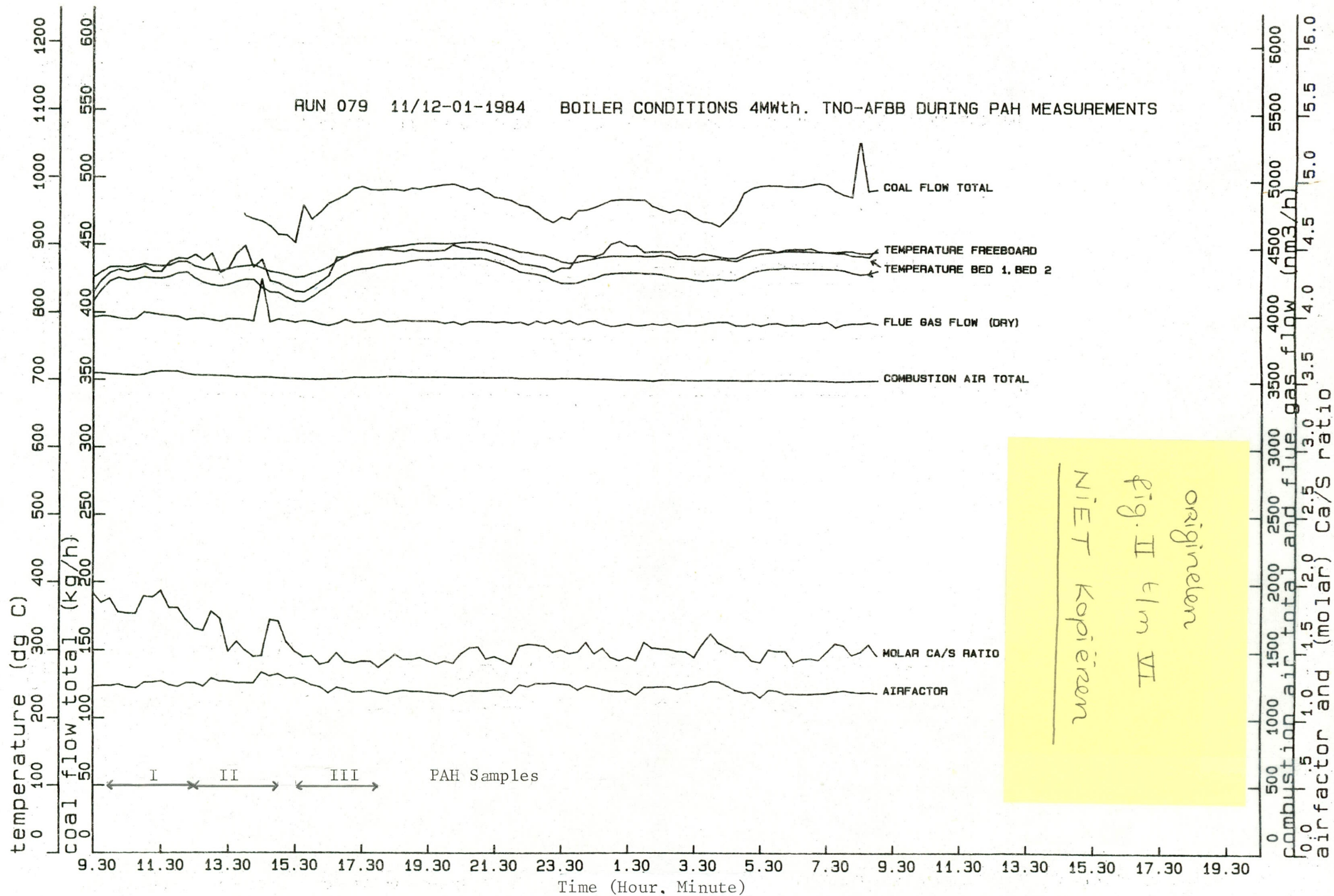




FIGURE II



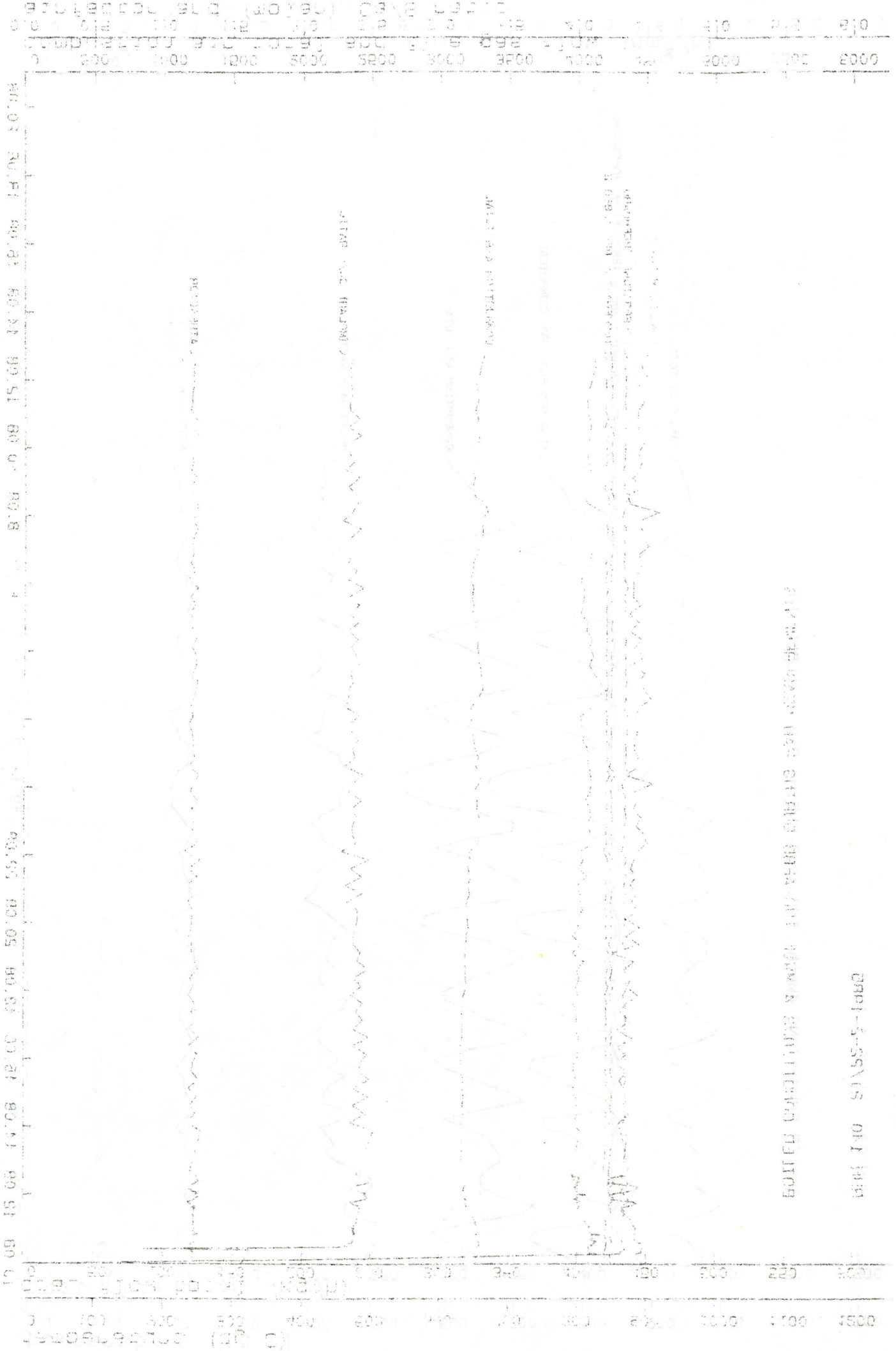
2. The second part outlines the various methods used to collect and analyze data, ensuring that the information gathered is reliable and valid. This includes both qualitative and quantitative approaches.

3. The third section details the results of the research, highlighting key findings and trends observed during the study. It provides a clear overview of the outcomes achieved.

4. Finally, the fourth part presents conclusions drawn from the analysis, along with recommendations for future research and practical applications based on the findings.

FIGURE I II

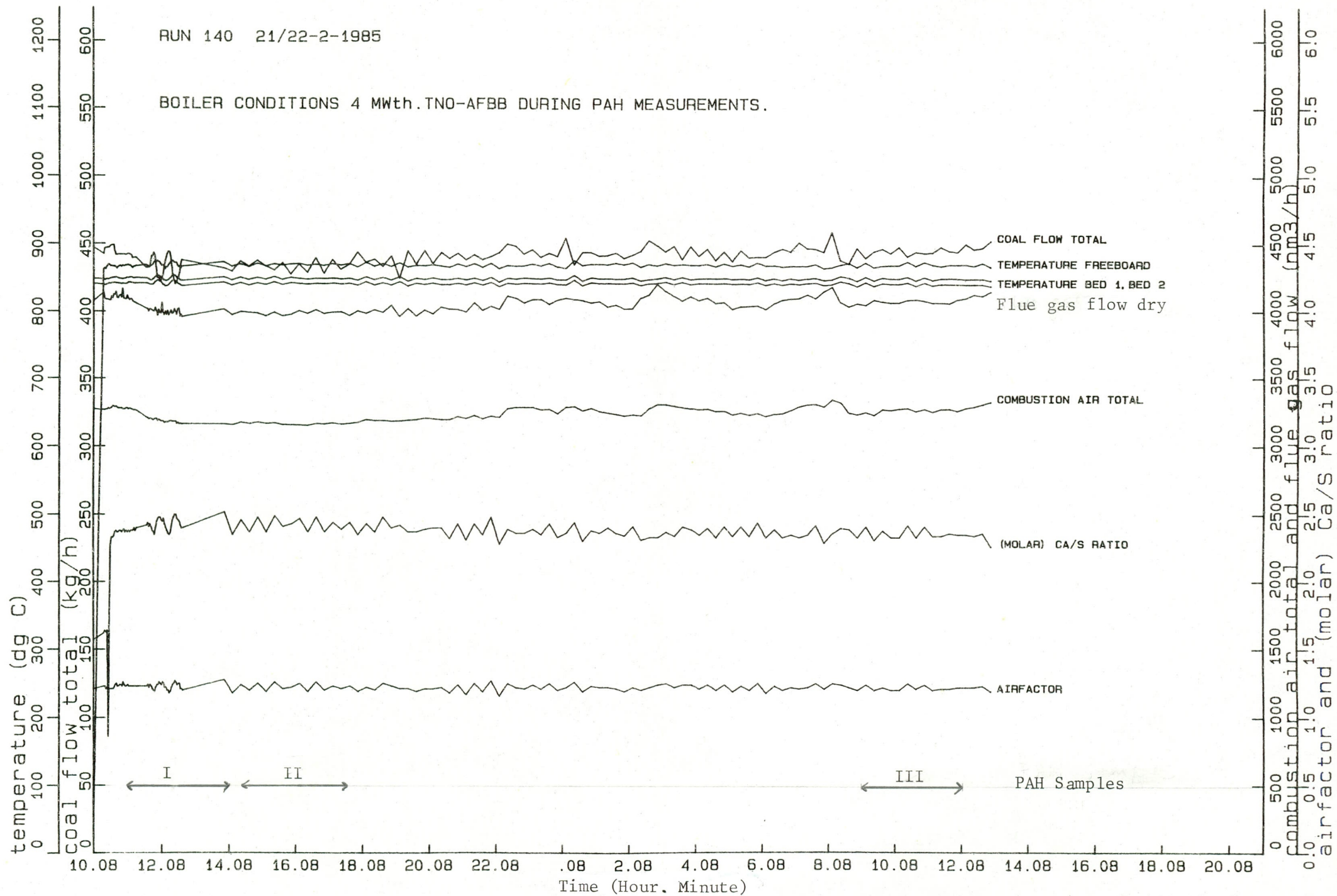




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FIGURE IV



7281-6-11-61 ENI 1000

STATIONARY POSITION, 10000 FT. ALTITUDE, 10000 FT. ALTITUDE



FIGURE V

