



HAL
open science

Factors influencing pollutant gas emissions of VOC recuperative incinerators : large-scale parametric study

Sylvain Salvador, Yilmaz Kara, Jean-Michel Commandre

► To cite this version:

Sylvain Salvador, Yilmaz Kara, Jean-Michel Commandre. Factors influencing pollutant gas emissions of VOC recuperative incinerators : large-scale parametric study. Applied Thermal Engineering, 2006, 26 (n° 14-15), p.1640-1651. 10.1016/j.applthermaleng.2005.11.025 . hal-01846928

HAL Id: hal-01846928

<https://hal.science/hal-01846928>

Submitted on 6 Nov 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Factors influencing pollutant gas emissions of VOC recuperative incinerators—Large-scale parametric study

S. Salvador ^{a,*}, Y. Kara ^b, J.-M. Commandré ^a

^a Ecole des Mines d'Albi-Carmaux, Centre Énergétique—Environnement, Laboratoire de Génie des Procédés des Solides Divisés, UMR CNRS 2392, Campus Jarlard, Route de Teillet, 81013 Albi CT Cedex 09, France

^b Gaz de France Research and Development Division, Industrial Utilisation Section, 361 Avenue du Président Wilson, 93211 Saint-Denis La Plaine Cedex, France

Abstract

This work establishes quantitative links between the operation parameters—plus one geometrical parameter—and the gas pollutant emissions of a recuperative incinerator (RI) of volatile organic compounds (VOCs). Using experimental design methodology, and based on a large number of experiments carried out on a half-industrial-scale pilot unit, mathematical expressions are established to calculate each of the pollutant emissions from the value of all the operation and design parameters. The gas emissions concerned are total hydrocarbons, and CO and NO_x emissions, while the control parameters are the flow rate of the treated air flow, the concentration of VOCs in the air flow, the preheating temperature of the flow, and the temperature at the exit of the combustion chamber. One design parameter—the aperture of the diaphragms—is also considered. We show that the constraining emissions are only that of CO and NO_x. Polynomials to predict them with a high accuracy are established. The air preheating temperature has an effect on the natural gas consumption, but not on CO and NO_x emissions. There is an optimal value for the aperture of the diaphragms, and this value is quantitatively established. If the concentration of VOCs in the air flow is high, CO and NO_x emissions both decrease and a high rate of efficiency in VOC destruction is attained. This demonstrates that a pre-concentration of VOCs in the air flow prior to treatment by RI is recommended.

Keywords: VOC; Recuperative incinerators; Pollutant emissions; Parametric study

1. Introduction

A large number of industrial sectors generate air flows contaminated with volatile organic compounds (VOCs). One can cite the printing industry, the perfume production industry and the chemical industry among the main sources resulting from human activity. Despite the fact that, on a global scale, natural sources account for 90% of these emissions, the situation is completely different in industrialised countries, where anthropic emissions exceed 80% of total VOC emissions. Transport usually accounts for more than

half of all anthropic emissions, but the use of organic solvents causes 30% of human-related emissions. VOCs are harmful to animals and plants, and they contribute to the greenhouse effect.

The present work is concerned with the elimination of VOCs resulting from industrial production, using one of the most standard techniques to destroy them when diluted in air flows: the recuperative incinerator (RI) [1,2]. This technique is an alternative to catalytic destruction of VOCs. The general principle, illustrated in Fig. 1, is to heat the air flux to a temperature of approximately 750 °C, and to maintain this temperature inside a Combustion Chamber (CC), typically for 0.5–1 s, in order to achieve an ideal and complete oxidation of VOCs into CO₂ and H₂O [3]. In practical terms, a destruction efficiency of 95–99.999% is

* Corresponding author. Tel.: +33 5 6349 3127/3027; fax: +33 5 6349 3243.

E-mail address: salvador@enstimac.fr (S. Salvador).

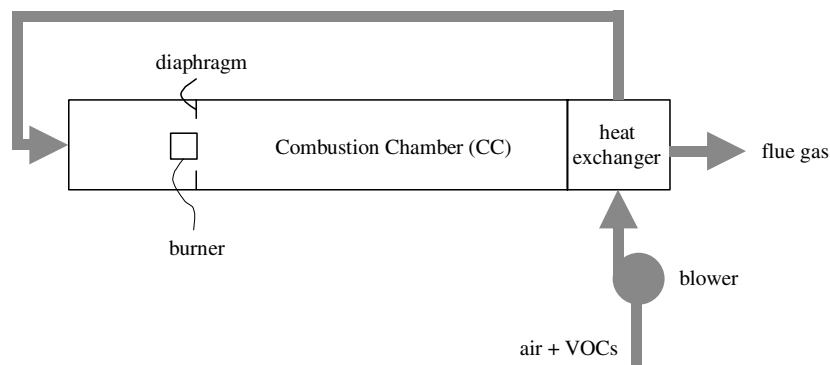


Fig. 1. General principle of a recuperative incinerator.

achieved using this technology. In most cases, the concentration of VOCs in the air flows to be treated by the process is not high enough to obtain the required energy to heat the air flux through the oxidation of the VOCs themselves. Consequently, a burner, usually fed with Natural Gas (NG), is used to supply this energy.

It is obvious that the lower the fuel consumption of the burner, the lower the cost of VOC destruction and the lower the CO₂ emissions of the process will be. Nevertheless, if reaction temperatures inside the CC are too low, this will result in a low destruction rate efficiency (DRE) of the VOCs, which in turn will lead to a high concentration of unburned hydrocarbons, or total hydrocarbon concentration (THC) in the flue gas of the RI. Operating at low combustion temperatures will also result in the emission of large quantities of carbon monoxide (CO). Moreover, one of the roles of the burner is to guarantee continuous and stable combustion of the VOCs. To this end, burners generate a “hard flame”: the temperatures inside the flame are reported to be in the range of 1000–2000 °C [4]. Consequently, a RI generates significant quantities of nitrogen oxide (NO); the higher the burner regime, the higher the NO emissions.

Summarising all this, a RI generates at least four pollutant gas species: CO₂, CO, THC and NO.

There are a number of parameters that can affect the gas emissions of a RI.

- The *reaction temperature* has opposite effects on the emissions of each pollutant: as we remarked before, an increase in the temperature will decrease CO and THC emissions, but will increase NO emissions.
- The *air-flow rate* through the RI will affect the residence time of VOCs inside the CC, and, in turn, the efficiency of the destruction of VOCs and of CO.
- In order to achieve high global thermal efficiency of the IR, part of the energy of the hot purified air flux out of the CC is recovered—using a gas/gas heat exchanger—to preheat the air plus the VOC flux. *Preheating temperatures* of 350–400 °C are typically reached. This parameter can potentially have an effect on pollutant gas emissions, and will be considered as a parameter in this work.

- High turbulence levels are created inside the CC; the main artifice is to put two diaphragms above and below the burner ramps—as will be discussed later (see Fig. 1). These diaphragms create high velocity air streams in the free section between the diaphragms and the wings of the burners, that we will call *air veins*. This enhances mixing of the hot gases from the burner’s flame with the air and VOC flux through the air veins, thanks to large recirculations downstream of each diaphragm. The *height of the air veins* is here considered as a design parameter.
- The *concentration in VOCs* and the nature of the VOCs can be very different from one industry to another. This concentration also can vary drastically over time in the same plant. Since a RI should be able to treat air flows with variable VOC concentrations, the concentration in VOC will be an additional parameter to consider.

In an industrial RI, the emissions of different pollutant gases are usually measured only occasionally. Parameters such as the air-flow rate and the VOC concentration tend to fluctuate, and are not known. Additionally, some parameters such as the CC temperature, the air-flow rate, and the preheating temperature are directly linked through the heat balances for the heat exchanger and the CC. Despite the very large number of RIs used around the world, there is, to date, no result that establishes quantitative links between the design, control and operating parameters and the emissions of pollutant gases. Today’s RIs do not always comply with regulations regarding all gaseous emissions, and there is a need to improve their design. These were the initial motivations of this study.

Given the multi-objective character of the problem, and the large number of parameters involved, it is very difficult to take a decision on a design improvement or to interpret the results from sporadic tests. The present work thus aims to provide guidelines for better control of an existing RI.

In this work, we have quantitatively established the impact of each of the design, control and operation parameters, taken separately—and this is the particularity of this work—on each of the pollutant gas emissions: CO, THC and NO (see below for CO₂). This work is based on an

important experimental study carried out using a large-scale finely instrumented RI pilot unit designed and built for this purpose. *Experimental design methodology* was used to minimise the number of experiments, which nevertheless reached a total of 63 for the RI, plus 42 experiments to characterise the burner alone, i.e., when the air flow does not contain VOCs.

2. Pilot unit design

The pilot unit is an exact replica, at half scale, of a standard industrial RI. The general principle of a VOC RI is recalled in Fig. 1; full details of the design of the CC can be found elsewhere [5,13]. The burner ramp consists of a NG distribution pipe and of two wings consisting of perforated refractory steel plates, as illustrated in Fig. 2. In the pilot unit, the air flux containing the VOCs was reconstituted by spraying liquid ethyl acetate ($C_4H_8O_2$) in the pre-heated air flux. The mounting to achieve this is detailed in Fig. 3. A calibrated pump was used to control the liquid VOC flow rate.

3. The experimental design technique

Experimental design methodology is a mathematical tool to help the experimenter to get the best knowledge of a process by running the minimum number of experiments. Detailed information about the various methods available can be found in the literature [6–10]. The NEM-ROD[®] software was used for this study. In this type of approach, the process is seen as a black box with outputs called *responses* or *objectives*. In the present case, the responses are, for instance, the different pollutant gas emissions. The responses may vary under the effect of *factors*, or *variables*. In our case, these might be the air-flow rate, the combustion temperature, the aperture of the air veins.

An experimental approach can go through different levels:

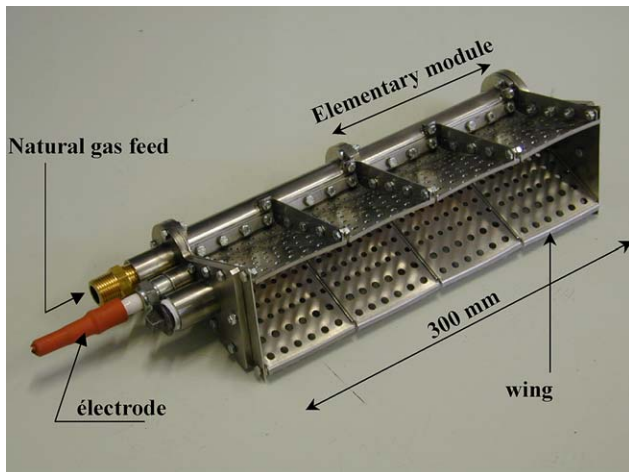


Fig. 2. Photograph of the burner—pilot unit.

- Screening: in this approach, the operator only wants to identify the parameters that have a significant effect on the responses.
- A quantitative study: here a mathematical expression is sought, that will enable the value of the response to be calculated from the values of the different parameters. These expressions will hereafter be called polynomials.
- Optimisation: The objective in this case is to determine the values of each parameter that minimises or maximises one or several responses. This is not treated in the present paper; an example for such a study can be found in [11].

For mathematical considerations, it is necessary to normalise the real variables, also called *natural variables*. Centred reduced variables, also called *reduced variables* X , are defined using the natural variables U :

$$\bar{U}_{ij} = \frac{U_{ij} - U_{0j}}{\Delta U_j}, \quad (1)$$

where i stands for the experiment index and j stands for the variable index. This requires defining a study domain for each of the variables. The centre of the domain is $U_{0,j}$, and the variable can range between $U_{0,j} - \Delta U_j$ and $U_{0,j} + \Delta U_j$. ΔU_j is called the variation step for the natural variable U_j . For instance the domain for the air-flow rate will be centred at $Q_{\text{air}} = 500 \text{ m}^3/\text{h}$ (at STP) and will vary by $100 \text{ m}^3/\text{h}$ (at STP) above and below this value.

The list of experiments to be carried out is determined through the construction of an experiment matrix. In a problem aiming to achieve a prediction capacity, an equiradial composite matrix is recommended. Equiradial means that the experimental points are located regularly on a circle, i.e., at the same distance from the centre of the experimental domains. The term composite means that the matrix is composed of three parts: a factorial matrix, a star matrix and several additional points at the centre of the domain.

Finally, an expression will be found to calculate each of the natural responses from the reduced variables, using a first degree linear model with interactions, i.e., of the type

$$U = b_0 + \sum_{i=1}^k b_i \bar{U}_i + \sum_{i=1}^{k-1} \sum_{j=i+1}^k b_{ij} \bar{U}_i \bar{U}_j.$$

A prediction coefficient can then be calculated. If the value is less than 70% [12], the polynomial is not accurate enough for predictions to be made.

4. The choice of factor ranges in the experimental design

The CC reaction temperature T_S is the control parameter *par excellence*. In an industrial application, it is maintained at a constant value—while the air-flow rate or VOC concentration might change—using classical thermocouple measurement and PID control. The range 700–

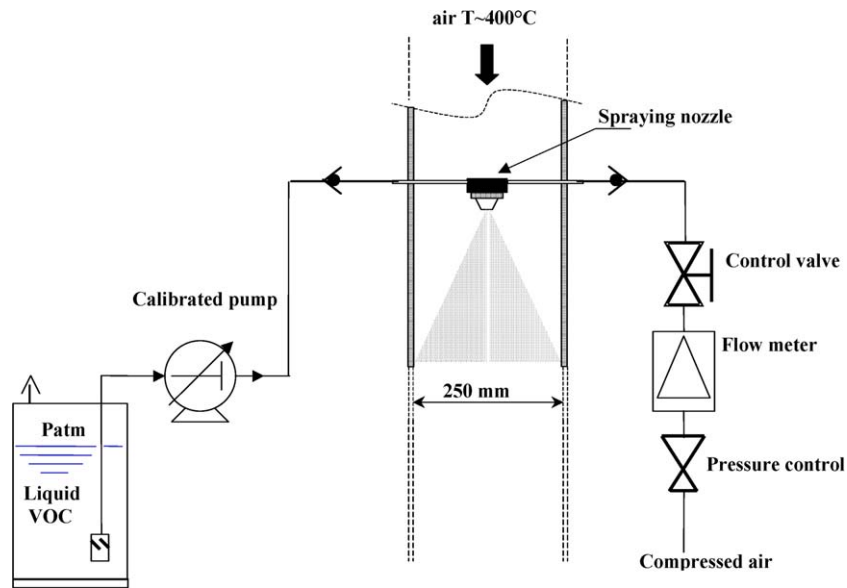


Fig. 3. Mounting for the reconstitution of the air and VOC flux.

750 °C—which covers situations from very low DRE and very high CO emissions to very high DRE and negligible CO emissions—was selected for this study. The CC temperature will act directly on the chemical reaction kinetics and only marginally on the gas residence time in the CC, given its low relative variation.

The air-flow rate Q_{air} can be seen as a design parameter. Indeed, during the design of a RI, the length of the burner ramp is adapted to the flow rate to be treated. Nevertheless, once an industrial RI is installed, the air-flow rate that it is required to treat might change over time; this flow rate then becomes an operation parameter. This parameter essentially acts on the average residence time of air in the CC. A high flow rate means low residence time, and consequently, low combustion efficiency. Additionally, the air-flow rate will impact on the turbulence level inside the CC, this time in favour of high combustion rates corresponding to high flow rates. We fixed the range 400–600 m³/h STP for our pilot unit, whose section is 320 mm wide by 360 mm high. This was determined to correspond with residence times ranging from 0.82 to 0.55 s, respectively, at an reaction temperature of 725 °C.

The concentration of VOCs of the air flux C_{VOC} is also a design parameter in the sense that, if it is known during the design of a RI, the geometry can be decided by taking account of this value. Again, this concentration can also vary with time during the operation of an installed industrial RI: this concentration then becomes an operation parameter. We adopted the range 4–8 mg/m³ STP, which corresponds to the standard domain for best operation of RI technology. As is the case for the air-flow rate, an industrial RI that is capable of treating air flows with a widely varying range of VOC concentrations will be an efficient RI.

The height of the air veins H_v is the only geometrical parameter considered in this study. It can be changed easily on a RI, but usually requires stopping the installation. The height of the air veins will impact on two main types of phenomena. Firstly, the diaphragms balance out the air-flow rates through the air veins and through the burner: decreasing the height of the air veins will force more air through the wings of the burner. This will affect the global fuel richness inside the flame zone of the burner. Second, the height of the diaphragms has a direct impact on the turbulence level inside the CC. This is because each diaphragm is an obstacle to the air flow; it will induce air recirculations downstream [4]. High diaphragms (or small air veins) induce high turbulence levels inside the CC. We adopted the range 30–40 mm for our half-scale pilot unit.

The air preheating temperature T_e , in the case of an industrial RI, results from the thermal efficiency of the *flue gas/fresh air* heat exchanger placed at the exit of the CC. This parameter was considered in this study in order to evaluate how far designing and paying for a high-efficiency heat exchanger was both cost-effective and environmentally positive. If the results indicate that high preheating temperatures are particularly interesting, the idea of preheating the air flux using an additional heat source could also be considered. The range 350–400 °C was adopted in this work.

A summary list of the parameters selected in the study and of their ranges can be found in Table 1.

5. The choice of responses in the experimental design

As previously stated, a RI will be the source of emissions of different gas species that are considered as pollutant gases.

Table 1
Factors and ranges for their values, for the experimental work to characterise the RI

		Natural variables		Reduced variables		Centre point	Variation step	In star matrix
		Expression	Value	Expression	Value			
Air flow rate	High level	Q_{air}	400 m ³ /h STP	$\bar{Q}_{\text{air}} = \frac{Q_{\text{air}} - 500}{100}$	-1	500 m ³ /h STP	100 m ³ /h STP	262 m ³ /h STP
	Low level		600 m ³ /h STP		1			
VOC concentration in air	High level	C_{VOC}	4 g COV/m ³ STP	$\bar{C}_{\text{COV}} = \frac{C_{\text{COV}} - 6}{2}$	-1	6 g COV/m ³ STP	2 g COV/m ³ STP	1.2 g COV/m ³ STP
	Low level		8 g COV/m ³ STP		1			
Air preheating temperature	High level	T_e	350 °C	$\bar{T}_e = \frac{T_e - 375}{25}$	-1	375 °C	25 °C	338 °C
	Low level		400 °C		1			
Combustion chamber output temperature	High level	T_s	700 °C	$\bar{T}_s = \frac{T_s - 725}{25}$	-1	725 °C	25 °C	668 °C
	Low level		750 °C		1			
Height of the air veins	High level	H_v	30 mm	$\bar{H}_v = \frac{H_v - 35}{5}$	-1	35 mm	5 mm	23.8 °C
	Low level		40 mm		1			

- VOCs, when submitted to high temperatures, will crack into lighter hydrocarbon species (HCs), following numerous and complex processes. These species will then be oxidised into both CO and CO₂, depending on local temperature levels, species concentrations and turbulence levels. Some of the HCs will not be oxidised. The remaining HC species will here be considered as a whole and called total hydrocarbons (THC). They will be given through an equivalent amount of C and expressed as an equivalent of CH₄. The measured ppm equivalent CH₄ have been converted to mg/m³ STP in this paper.
- Similarly, part of the CO formed as an intermediate species will not be oxidised to CO₂ and will be considered in this work. The rejected amounts will be expressed both in mg/m³ STP and through the CO emission index (COEI). The COEI is the ratio of the CO mass flow rate to the treated VOC and natural gas mass flow rate.
- A RI also generates NO_x. Natural gas is used as the auxiliary source of energy in the process. Neither VOCs nor NG contain significant quantities of N. Consequently, since prompt NO is not formed in the RI process [4,13], NO emitted by a RI is *Thermal NO_x*. The amounts released will also be expressed both in mg/m³ STP and through the NO emission index (NO_xEI). The NO_xEI is the ratio of the NO_x mass flow rate to the treated VOC and natural gas mass flow rate.
- The oxidation of the VOCs and of the NG will largely result in the emission of CO₂, which undoubtedly has an environmental impact. Nevertheless, the emissions of this species will not be considered here since its production is directly linked to the amount of VOC that is oxidised, and to the quantity of NG that is used to achieve this goal. Natural gas consumption is taken into account in this study via the examination of the CC reaction temperature. A simple heat balance for the RI will give the link between the NG consumption, the VOC flow rate and CO₂ emissions.

For most of the pollutant gas emissions considered here, regulatory emission limits have been established. We will

refer to the French regulations [14]. The maximum values are 100 mg/m³ STP for CO emissions, 1000 mg/m³ STP for NO emissions, and 20 mg/m³ STP for THC emissions.

6. Results and discussion

6.1. Pre-estimation of the sensitivity of the gas emissions to the selected factors

As a first step, the screening study was carried out, based on the results of a limited number of experiments, following the complete factorial matrix, in order to gain an initial estimation of the impact of each factor on each response. This matrix is given in Table 2a, together with the responses obtained for each of the 32 experiments. The factors are expressed using real variables.

The results obtained are presented in a semi-quantitative way in Table 3. A “+” sign corresponds to a 5% increase in the response (centred on the global effect b_0) for an increase in the considered factor from the level -1 to the level +1 in coded variables. A “-” sign corresponds to an equivalent decrease in the response. It should be remembered that these effects are characterised around a central point, where all the coded variables equal zero (*centre of the domain* column in Table 1).

An examination of the results in Table 2a shows that the emissions of all the three pollutants considered here vary in wide ranges. The emissions of CO range from less than 40 ppm in experiments 9, 13 and 15 to several thousands ppm, well above the regulation values. The emissions in unburned HC reach 112 mg CH₄/m³ at STP in experiment 20, corresponding with a destruction rate efficiency of only 97.4%. On the other hand, most of the time, the THC emissions are below the detection limit of the analyser, i.e., 10 ppm, corresponding to a *DRE* better than 99.8%. The emissions in NO_x vary from 21.6 to 92.7 mg/m³ at STP, where the regulation limit is 100 mg/m³ STP. These results confirm the need for good design and control of the RI process in order to limit the environmental impact of the operation. The four parameters Q_{air} , C_{VOC} , T_s and H_v have strong effects on the different gas emissions. The effect

Table 2
Factors and responses for the RI

No. Exp.	Natural variables					Response						
	Q_{air} (m ³ /h)	C_{VOC} (g COV/m ³)	T_e (°C)	T_s (°C)	H_v (mm)	CO (mg/m ³)	NO _x (mg/m ³)	THC (mg/m ³)	NG (l/min)	COEI (g/kg _{fuel})	NO _x EI (g/kg _{fuel})	Power (kW)
(a)												
1	400	4	350	700	30	2183	59.7	1	95.0	154.0	4.4	55.4
2	600	4	347	700	30	2256	39.7	36	141.5	160.0	2.94	82.5
3	400	8	350	700	30	3806	44.8	1	80.0	229.7	2.83	46.7
4	600	8	348	700	30	4275	27.3	43	120.0	258.0	1.72	70.0
5	400	4	400	700	30	2356	56.8	1	85.0	179.8	4.53	49.6
6	600	4	401	700	30	2314	35.9	36	132.0	172.3	2.8	77.0
7	400	8	400	700	30	3513	32.4	1	71.0	225.1	2.17	41.4
8	600	8	401	700	30	4406	21.6	43	108.0	280.4	1.44	63.0
9	400	4	350	751	30	39	81.7	1	109.0	2.5	3.8	63.6
10	600	4	350	750	30	275	58.9	1	156.0	18.2	3.12	91.0
11	400	8	350	750	30	49	64.4	1	90.0	2.8	2.73	52.5
12	600	8	354	750	30	413	40.7	1	132.0	23.4	2.01	77.0
13	400	4	400	751	30	39	84.1	1	98.2	2.7	4.3	57.3
14	600	4	400	750	30	264	56.2	1	144.0	18.5	3.22	94.0
15	400	8	400	750	30	38	62.7	1	81.0	2.3	2.77	47.2
16	600	8	400	750	30	525	37.1	1	117.0	32.1	1.97	68.2
17	400	4	350	701	40	2050	64.2	39	97.0	142.5	4.61	56.6
18	600	4	350	702	40	2038	61.5	72	144.0	142.7	4.5	84.0
19	400	7.5	350	700	40	3613	56.2	21	83.3	220.0	3.58	48.6
20	600	8	352	700	40	3950	47.1	112	120.1	238.3	2.97	70.0
21	400	4	405	700	40	2275	66.2	29	85.5	172.9	5.18	49.9
22	600	4	394	700	40	2194	62.1	66	128.6	166.4	4.92	75.0
23	400	8	405	700	40	3625	53.0	18	71.1	232.1	3.5	41.5
24	600	8	393	700	40	4275	49.1	93	105.3	275.5	3.31	61.4
25	400	4	350	750	40	225	86.8	1	107.5	14.5	4.24	62.7
26	600	4	352	750	40	638	81.9	1	159.1	41.5	4.38	92.8
27	400	8	351	750	40	188	71.3	1	90.5	10.6	3	52.8
28	600	8	353	750	40	1056	65.6	1	129.1	61.3	3.21	75.3
29	400	4	405	750	40	131	92.7	1	98.1	9.1	4.85	57.2
30	600	4	402	750	40	488	87.4	1	145.5	33.9	4.83	84.9
31	400	8	402	750	40	169	72.7	1	80.1	10.2	3.31	46.7
32	600	8	400	750	40	700	65.8	1	116.7	42.9	3.24	68.1
(b)												
33	262	6	375	726	35	109	61.7	1	60.5	6.8	3.1	35.3
34	738	6	376	724	35	2813	45.4	24	168.0	178.5	3.01	98.0
35	500	1.2	375	725	35	798	74.6	1	132.0	63.5	5.62	77.0
36	500	10.8	374	726	35	2675	37.3	4	83.0	149.7	2.12	48.4
37	500	6	338	724	35	1365	53.8	1	112.0	87.5	3.23	65.3
38	500	6	435	723	35	1300	57.0	1	96.0	91.4	3.66	56.0
39	500	6	377	668	35	3691	38.1	197	97.0	257.9	2.78	56.6
40	500	6	376	784	35	20	87.2	1	129.0	1.2	3.6	75.2
41	500	6	375	725	23.8	1945	37.0	1	107.2	211.8	4.0	62.5
42	500	6	376	725	46.2	1948	76.0	25	110.3	206.1	8.0	64.3
43	500	6	376	725	35	1285	57.0	1	108.0	84.2	3.31	63.0
44	500	6	375	725	35	1271	57.8	1	108.0	83.3	3.38	63.0
45	500	6	375	727	35	1175	60.3	1	108.0	77.0	3.55	63.0
46	500	6	375	725	35	1303	58.5	1	108.0	85.4	3.44	63.0
47	500	6	375	725	35	1284	55.4	1	108.0	84.2	3.37	63.0
48	500	6	376	724	35	1368	57.9	1	108.0	89.6	3.55	63.0
49	500	6	376	725	35	1289	56.4	1	108.0	84.5	3.36	63.0
50	500	6	374	724	35	1365	64.0	2	108.0	89.5	3.81	63.0
51	500	6	375	722	35	1556	56.6	3	108.0	102.0	3.44	63.0
52	500	6	375	724	35	1369	59.1	3	109.0	89.2	3.66	63.6
53	400	8	400	700	30	3388	32.4	1	71.0	217.1	2.17	41.4
54	400	4	405	750	30	31	78.8	1	98.2	2.2	4.03	57.3
55	400	8	351	750	40	263	73.1	1	88.5	15.0	3.26	51.6
56	500	6	377	726	40	1700	66.6	6	108.0	111.4	3.99	63.0
57	300	6	375	726	40	575	71.9	1	74.8	34.5	3.54	43.6
58	500	6	375	675	40	3400	55.0	171	99.9	167.8	2.84	58.3

(continued on next page)

Table 2 (continued)

No. Exp.	Natural variables					Response						
	Q_{air} (m ³ /h)	C_{VOC} (g COV/m ³)	T_e (°C)	T_s (°C)	H_v (mm)	CO (mg/m ³)	NO _x (mg/m ³)	THC (mg/m ³)	NG (l/min)	COEI (g/kg _{fuel})	NO _x EI (g/kg _{fuel})	Power (kW)
59	500	6	375	726	40	1369	75.4	1	112.7	87.4	4.22	65.7
60	500	2	376	726	40	988	84.9	7	128.2	76.1	6.13	74.8
61	500	6	375	725	40	1438	72.5	1	109.0	93.7	4.32	63.6
62	500	10	375	726	40	2188	56.8	1	90.0	123.5	2.96	52.5
63	500	6	375	725	40	1550	70.5	1	109.0	101.0	4.16	63.6

(a) Complete factorial plan. (b) Complementary experiments, including the repetition experiments at the centre of the domain (43–52), and the preliminary experiments (53–63); power indicated corresponds to the energy supplied by natural gas only.

Table 3
Semi-quantitative sensitivities of the responses to the factors

	NO _x	CO	THC
Q_{air}	--	++	++
C_{VOC}	-----	++++	
T_e			
T_s	++++	-----	-----
H_v	++++		++
b_0	31	1258	9

of a given factor can be either positive or negative depending on the pollutant species that is considered, as is the case for the parameters Q_{air} , C_{VOC} and T_s . In the case of H_v , the results indicate an increase in both NO_x and THC emissions when H_v is increased. It seems that the preheating temperature T_e has no impact on the gas emissions. One should not forget that these effects are established from the variation of the responses using only two values for a given factor: the minimum and the maximum. As a consequence, a bell-shaped response can be misinterpreted as a value zero at this stage. One can nevertheless conclude that at least four out of the five factors have a strong effect on the responses, but a more careful characterisation of the sensitivities is needed. This is done in the following section on determination of polynomial expressions.

6.2. Determination of the polynomial expressions to predict gas emissions

The second step consists in determining polynomial expressions that predict each of the responses, or gas emissions, from the values of the factors, or parameters. This requires more numerous experimental data. Ten complementary experiments were carried out following the star matrix; the experimental conditions and the gas concentrations obtained are reported in Table 2b, experiments 33 to 42. Ten repetition experiments at the centre of the domain were also carried on in experiments 43 to 52. Finally, the results from preliminary experiments –53 to 63—also carried out during the project, were integrated in the data.

The coefficients for the three polynomial expressions were determined using multiple regression. The expressions are given in Eqs. (2)–(4) using coded variables. These emis-

sions can be expressed from real variables using the expression for the coded variables given in Table 1:

THC (mg CH₄/N m³)

$$\begin{aligned}
&= 6.508 + 9.754\bar{Q}_{\text{air}} + 1.331\bar{C}_{\text{VOC}} - 2.299\bar{T}_e \\
&\quad - 24.826\bar{T}_s + 9.001\bar{H}_v + 0.417\bar{Q}_{\text{air}}^2 - 1.564\bar{C}_{\text{VOC}}^2 \\
&\quad + 0.291\bar{T}_e^2 + 12.916\bar{T}_s^2 + 0.165\bar{H}_v^2 + 0.093\bar{Q}_{\text{air}}\bar{C}_{\text{VOC}} \\
&\quad - 0.146\bar{Q}_{\text{air}}\bar{T}_e - 0.528\bar{C}_{\text{VOC}}\bar{T}_e - 7.066\bar{Q}_{\text{air}}\bar{T}_s \\
&\quad - 7.191\bar{C}_{\text{VOC}}\bar{T}_s + 1.049\bar{T}_e\bar{T}_s + 0.716\bar{Q}_{\text{air}}\bar{H}_v \\
&\quad + 2.174\bar{C}_{\text{VOC}}\bar{H}_v - 5.515\bar{T}_e\bar{H}_v - 7.721\bar{T}_s\bar{H}_v, \quad (2)
\end{aligned}$$

CO (mg CO/N m³)

$$\begin{aligned}
&= 1455.5 + 290.2\bar{Q}_{\text{air}} + 474.9\bar{C}_{\text{VOC}} - 14.07\bar{T}_e \\
&\quad - 1195.9\bar{T}_s + 0.788\bar{H}_v + 6.063\bar{Q}_{\text{air}}^2 - 9.863\bar{C}_{\text{VOC}}^2 \\
&\quad - 1.675\bar{T}_e^2 + 55.65\bar{T}_s^2 + 122.9\bar{H}_v^2 + 94.78\bar{Q}_{\text{air}}\bar{C}_{\text{VOC}} \\
&\quad - 1.050\bar{Q}_{\text{air}}\bar{T}_e - 2.800\bar{C}_{\text{VOC}}\bar{T}_e + 26.388\bar{Q}_{\text{air}}\bar{T}_s \\
&\quad - 403.4\bar{C}_{\text{VOC}}\bar{T}_s - 8.338\bar{T}_e\bar{T}_s + 18.9630\bar{Q}_{\text{air}}\bar{H}_v \\
&\quad + 7.400\bar{C}_{\text{VOC}}\bar{H}_v - 6.738\bar{T}_e\bar{H}_v + 87.30\bar{T}_s\bar{H}_v, \quad (3)
\end{aligned}$$

NO_x (mg NO_x/N m³)

$$\begin{aligned}
&= 61.135 - 6.038\bar{Q}_{\text{air}} - 9.508\bar{C}_{\text{VOC}} - 0.575\bar{T}_e \\
&\quad + 11.37\bar{T}_s + 9.837\bar{H}_v - 1.068\bar{Q}_{\text{air}}^2 + 1.417\bar{C}_{\text{VOC}}^2 \\
&\quad + 0.226\bar{T}_e^2 + 1.027\bar{T}_s^2 - 1.109\bar{H}_v^2 + 0.493\bar{Q}_{\text{air}}\bar{C}_{\text{VOC}} \\
&\quad - 0.308\bar{Q}_{\text{air}}\bar{T}_e - 0.780\bar{C}_{\text{VOC}}\bar{T}_e - 0.596\bar{Q}_{\text{air}}\bar{T}_s \\
&\quad - 0.390\bar{C}_{\text{VOC}}\bar{T}_s + 0.513\bar{T}_e\bar{T}_s + 3.409\bar{Q}_{\text{air}}\bar{H}_v \\
&\quad + 1.273\bar{C}_{\text{VOC}}\bar{H}_v + 0.411\bar{T}_e\bar{H}_v + 0.021\bar{T}_s\bar{H}_v. \quad (4)
\end{aligned}$$

The prediction coefficients for the polynomials that predict CO and NO_x emissions are better than 92%, indicating that the predictions can satisfactorily be trusted [12]. The regression coefficient for the polynomial that predicts THC emissions is small at 59%. This is simply due to the fact that the data values for this species are most of the time at zero. As we will show later on, THC emission is not the restricting response for the RI process; the predicting polynomial will only be used in the remainder of the paper to make a qualitative analysis of the impact of the factors on this gas emission. We show in Fig. 4 a compar-

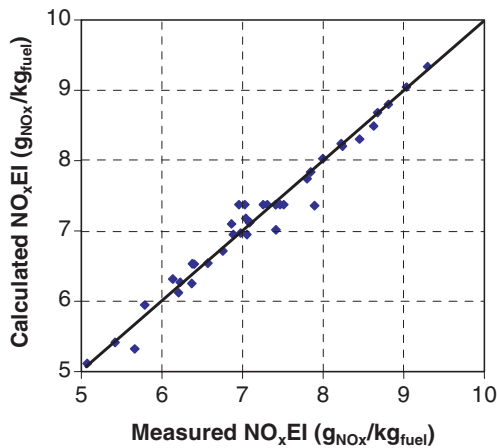


Fig. 4. Comparison between the experimental values for NO_xEI and the values calculated from the polynomial.

ison between the experimental NO emissions and the values predicted by the polynomial. This attests to the quality of the predictability of the polynomial.

6.3. Parametric study

The polynomial expressions in Eqs. (2)–(4) are here used to plot the trend in each of the pollutant gas emissions versus each design, control or operation parameter. The operating parameter C_{VOC} will be used as an additional plot parameter in the figures. The parameters are varied one at a time, while—for simplicity's sake—the other parameters are fixed at the centre point (see Table 1) of the experimental domain.

6.3.1. Influence of the temperature inside the combustion chamber

We have plotted in Fig. 5 the THC concentration at the exit of the CC versus the reaction temperature, for the different initial concentrations of VOC in the air flux. The trend observed is as expected, with THC concentrations decreasing when the temperature is increased. The regulation limit of $20 \text{ mg/m}^3 \text{ STP}$ is reached when the temperature increases beyond $700\text{--}715 \text{ }^\circ\text{C}$. HCs are totally oxidised for a temperature of $730 \text{ }^\circ\text{C}$ whatever the concentration of VOC in the air flux, the larger the emissions of THC. Nevertheless, if one calculates the DRE at a given temperature—say $680 \text{ }^\circ\text{C}$ —one finds values of 0.970, 0.981, 0.984, 0.987 and 0.989 for VOC concentrations in the air of 2, 4, 6, 8 and $10 \text{ mg/m}^3 \text{ STP}$, respectively. This result indicates that high initial VOC concentrations lead to high DRE. This result is important if one considers an industrial application of the process, and could justify the use of VOC pre-concentration techniques upstream of a RI. Indeed, the higher the DRE, the lower the operation temperature required and the lower the CO_2 emissions. This will nevertheless be relevant only in the case where the aim is to obtain very high DREs. In the present case where a DRE

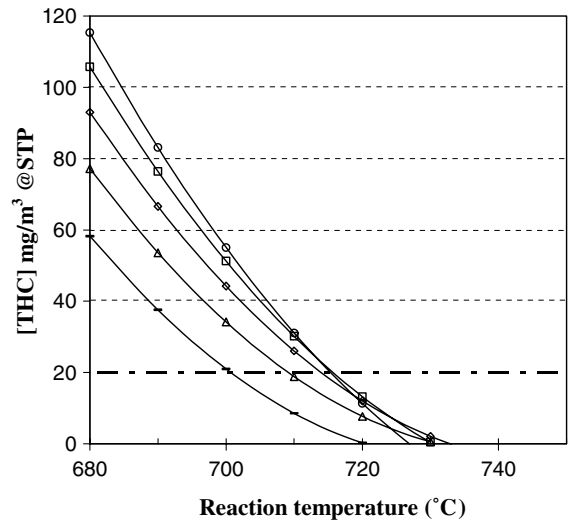


Fig. 5. Influence of the exit temperature of the combustion chamber on the emissions of THC for different initial concentrations of VOC in the air flux.

of 99.4% is sufficient ($\text{THC} < 20 \text{ mg/m}^3 \text{ STP}$ for an initial VOC concentration of $6 \text{ g/m}^3 \text{ STP}$), the minimum temperature is set by the maximum amount of CO at the exit, and not by the DRE, as we show below.

The emissions of CO , as illustrated in Fig. 6, follow the same general trends, again indicating high sensitivities both to the reaction temperature and to the VOC concentration in the air flux. This time, a higher temperature, i.e., $755 \text{ }^\circ\text{C}$, must be reached for the CO emissions to fulfil regulation emission limits of $100 \text{ mg/m}^3 \text{ STP}$, and this whatever the initial VOC concentration in the air flux. This comparison of minimum temperatures to destroy CO and HC is particularly important: it shows that the more constraining target is CO oxidation and not VOC or HC elimination.

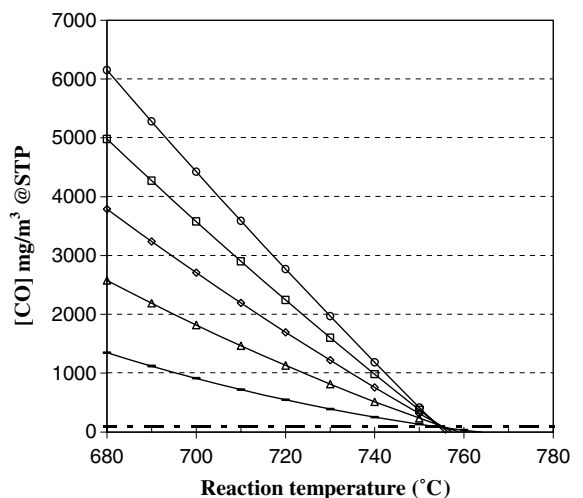


Fig. 6. Influence of the exit temperature of the combustion chamber on the emissions of CO for different initial concentrations of VOC in the air flux: (\circ) 2 g/m^3 , (\square) 4 g/m^3 , (\diamond) 6 g/m^3 , (\triangle) 8 g/m^3 , (—) 10 g/m^3 , (---) maximum regulation value.

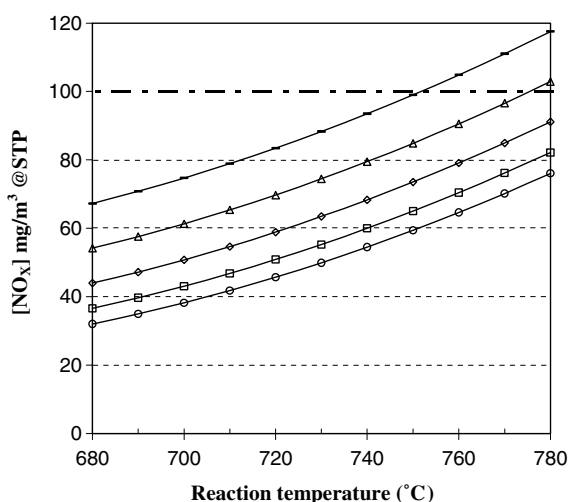


Fig. 7. Influence of the exit temperature of the combustion chamber on the emissions in NO_x for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

If the temperature is decreased by 50 °C only, CO fractions as high as several thousands of ppm are released. Moreover, the higher the initial VOC concentration, the higher the CO emissions, with an approximately linear trend.

The emissions of NO are very sensitive to the CC temperature and also to the initial VOC concentration in the air flux. Indeed, as indicated by the results plotted in Fig. 7, the NO fraction in the flue gas can range from 33 to 116 mg/m³ STP depending on these parameters (for an air-flow rate of 500 m³/h STP). High CC temperatures lead to high emissions of NO. This can be simply explained by the fact that high temperatures require a high regime for the burner, which is known to generate more NO_x [4,15].

When the initial concentrations of VOC in the air flux are high, the quantity of NO_x released is lower. This can be explained as follows: when high amounts of VOC are fed into the air flux, the burner regime is lower, for a given CC temperature. For instance, the energy brought by 6.9 mg/m³ STP in the air flux represents 35% of the energy for self-combustion. Since it is the burner that generates NO_x, as will be shown later, the emissions are lower when the concentration of VOCs increases. For the higher temperatures studied here and for the lower VOC concentrations, the emissions in NO_x increase beyond the regulation limit of 100 mg/m³ STP.

This result indicates that there is an advantage in creating a pre-concentration of VOCs in the air flux before treatment by a RI. Indeed, the elimination of a given quantity of VOC will lead to a smaller global quantity of NO released.

6.3.2. Influence of the air and VOC flow rate

As far as THC emissions are concerned, the results in Fig. 8 show a general increase when the flow rate is

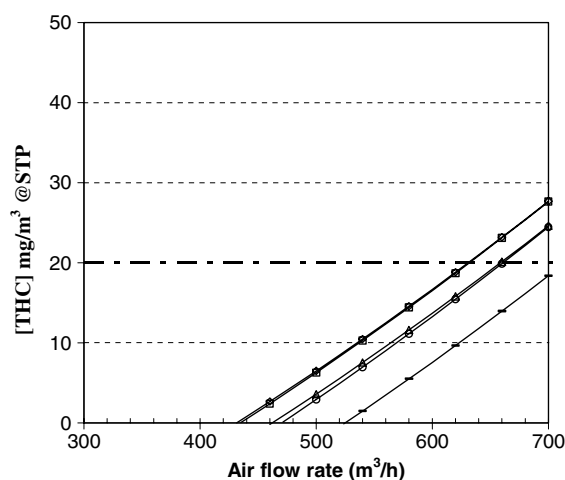


Fig. 8. Influence of the air-flow rate on the emissions of THC for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

increased. The regulation limit of 20 mg/m³ STP can rapidly be breached if the flow rate is too high. Flow rates smaller than 430 m³/h STP lead to the total destruction of VOCs (for a CC temperature of 725 °C).

Around the central point chosen for these plots, the emissions in CO are well above the regulation limits, as indicated by Fig. 9. This has the advantage of enabling the parameters influencing CO emissions to be clearly studied. The emissions of CO increase proportionally to the air-flow rate.

The impact of the air-flow rate on the NO_x emissions can be analysed from Fig. 10. An increase in the flow rate is globally accompanied by a decrease in the NO emissions. The explanation for this is not immediately apparent. One might expect that an increase in the flow rate, which will require an increase in the burner regime in order to keep

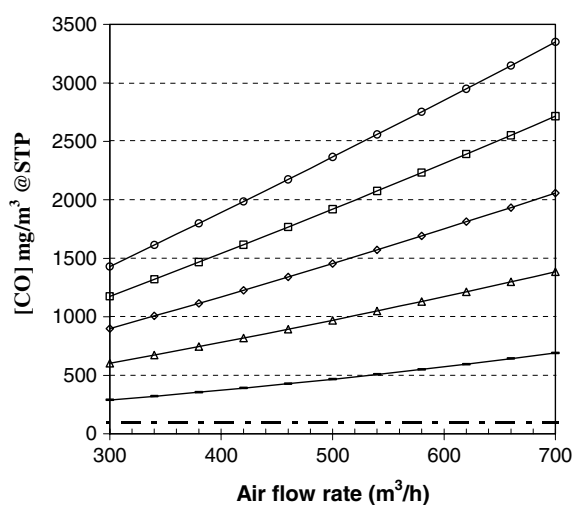


Fig. 9. Influence of the air-flow rate on the emissions of CO for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

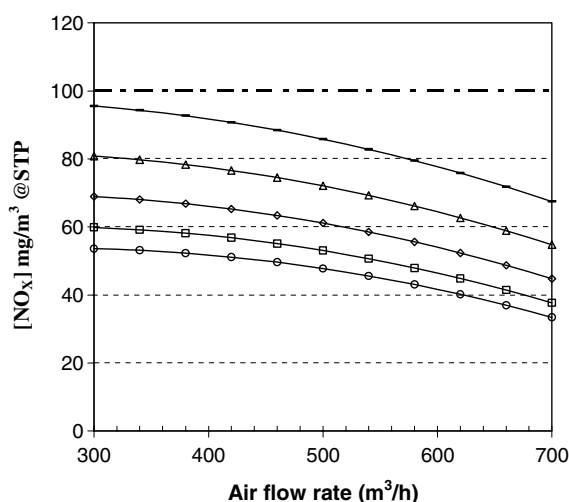


Fig. 10. Influence of the air-flow rate on the emissions of NO for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

a constant CC temperature, would be the source of higher NO_x emissions. An explanation for the present result is, in our view, that high air-flow rates induce lower global fuel richness inside the flame of the burner, whose temperature will decrease: NO_x formation will be limited. See our previous work [13].

6.3.3. Influence of the height of the air veins

We shall now focus on the design parameter *height of the air veins*, H_v , and assess its effect on the pollutant gas emissions. From the results plotted in Fig. 11, increasing the aperture of the air veins increases the emissions of THC. It is difficult to interpret this response. One explanation may be that increasing H_v reduces the turbulence level inside the CC, which could be at the origin of the lower efficiency of HC oxidation inside the CC.

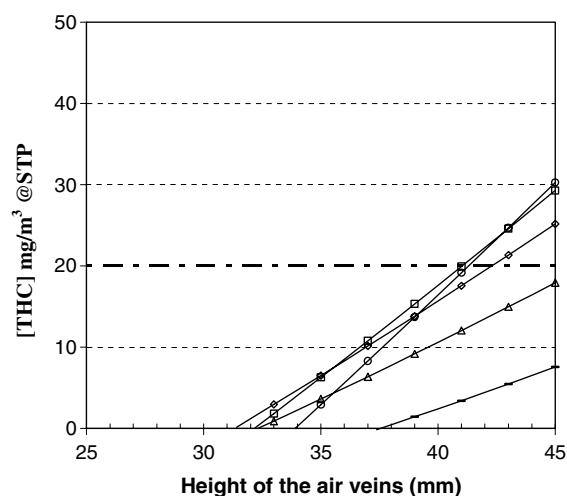


Fig. 11. Influence of the height of the air veins on the emissions of THC for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

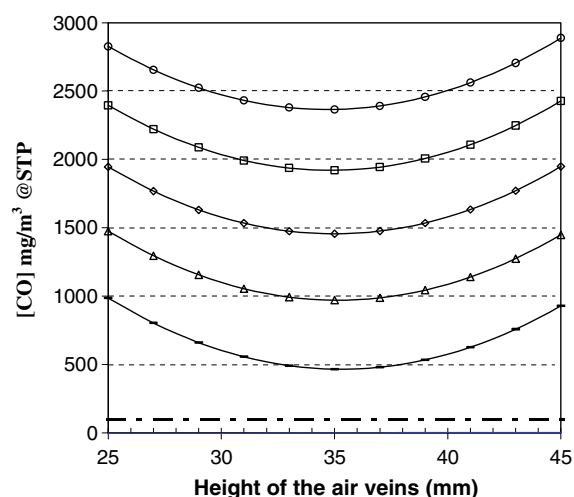


Fig. 12. Influence of the height of the air veins on the emissions of CO for different initial concentrations of VOC in the air flux: (○) 2 g/m³, (□) 4 g/m³, (◇) 6 g/m³, (△) 8 g/m³, (—) 10 g/m³, (- - -) maximum regulation value.

Of particular interest is the examination of CO emissions when H_v is varied. Fig. 12 shows that an optimum value exists for H_v , where the emissions in CO are at a minimum. This value is 35 mm in our pilot unit, corresponding to an average velocity of 14.5 m/s through the air veins (from CFD modelling). The emissions in CO are very sensitive to this parameter: a change of 10 mm can double the emissions. The bell shape of the curves suggests the presence of two antagonistic mechanisms. As explained before, increasing H_v decreases the turbulence level inside the CC, decreasing the efficiency of CO oxidation into CO₂. On the other hand, increasing H_v decreases the air-flow rate through the burner. The temperature of the flame is then likely to be higher—due to the increase in the global fuel richness—and the residence time inside the flame also higher, two factors that will favour the oxidation of CO inside the flame zone.

The emissions of NO_x are also very sensitive to H_v . Fig. 13 clearly indicates an increase in NO emissions when H_v is increased; a change from 25 to 45 mm in H_v induces an increase of NO from 18 to 68 mg/m³ STP when the initial VOC concentration in the air is 2 g/m³, and from 67 to 96 ppm when the initial VOC concentration in the air is 10 g/m³. When H_v is increased, the air-flow rate through the burner decreases, and the global fuel richness of the burner increases: the flame temperature increases and NO emissions follow. The increase in H_v is also accompanied by less oxidant conditions inside the flame zone, which should lead to lower NO emissions. It seems that this last phenomenon is less relevant than the effects of temperature.

6.3.4. Influence of the air preheating temperature

The polynomial expression confirms the indication of the previous screening study: the preheating temperature, when varied in the range 350–400 °C, does not affect the

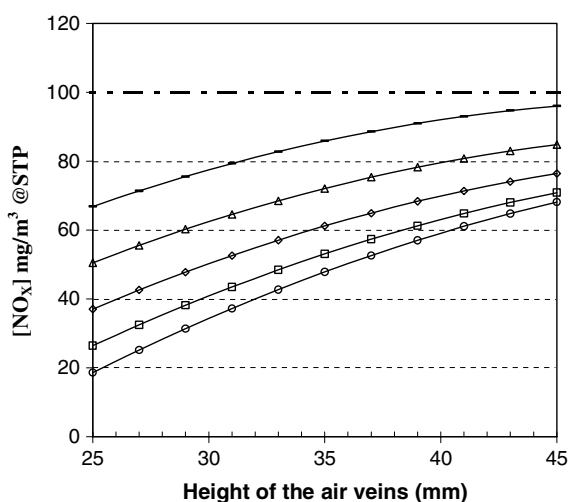


Fig. 13. Influence of the height of the air veins on the emissions of NO_x for different initial concentrations of VOC in the air flux: (○) 2 g/m^3 , (□) 4 g/m^3 , (◇) 6 g/m^3 , (△) 8 g/m^3 , (—) 10 g/m^3 , (- - -) maximum regulation value.

gas emissions significantly. The emissions in THC, CO and NO vary in the ranges 9–4.5, 1468–1440 and 62–61, respectively, when this temperature is varied from 350 to 400 °C. The implications of this result are as follows. First, it means that the thermal efficiency of the flue-gas/air-flux heat exchanger will only impact on the CO_2 emissions, via the global thermal efficiency of the RI, and not on the other pollutant gas emissions. It also indicates that there is no indication to preheat the air flow using an external heat source in order to limit pollutant gas emissions.

7. Case of the burner operated without VOC

The previous results lead us to distinguish between two types of pollutants: those originating from the burner and those originating from the combustion of VOCs that do not cross the wings of the burner, i.e., crossing the air veins. In order to obtain reliable results following this interpretation, a series of experiments was carried out in order to quantify the gas emissions originating from the burner alone. This was achieved by following a new experimental study in which no VOC was injected in the air flux. For simplicity's sake, details will not be given here. The results from this study clearly showed that running the burner alone does not result in any emission, either of THC or of CO. This shows that the full oxidation of CH_4 into CO_2 and H_2O is achieved. Consequently, in a RI, HC and CO emissions result from the combustion of the VOCs alone, and not from the NG supply.

Additionally, the NO_x EI registered for the burner alone and for the RI while incinerating the VOCs indicated that NO_x are produced only by the combustion of the NG, and not by the combustion of VOCs. This closely corroborates the previous observation that increasing the VOC concen-

tration—and consequently decreasing the NG flow rate—lead to a decrease in NO_x emissions.

8. Conclusion

A significant experimental study, organised following the *experimental design methodology*, was successfully carried out using a well-instrumented industrial-scale recuperative incinerator. It was possible to determine quantitatively the sensitivities of each of the pollutant gas emissions CO, NO_x and THC to each design, operation or control parameter separately: the combustion chamber temperature, air-flow rate, air veins aperture and the air preheating temperature. This sensitivity was established over a wide range for each parameter, and for concentrations of VOCs in the air flux ranging from 2 to 10 g/m^3 STP.

The air-flux preheating temperature does not have a significant impact on any of the pollutant gas emissions studied here: CO, THC and NO. This parameter will nevertheless have an impact on NG consumption and CO_2 emissions, as it directly affects the global thermal efficiency of the RI.

The three parameters *reaction temperature*, *air-flow rate* and *height of the air veins* have strong effects on all three pollutant gas emissions. An increase in the value of one parameter always results in an increase in the emissions of several pollutants and by a decrease in the emissions of others. The data obtained here will help in the control of an existing RI, since the impact of a given parameter has been characterised. The results also indicate that, depending on the operating conditions (air-flow rate, concentration of VOCs in air), an optimal value exists for the design parameter *height of the air veins* and for the CC output temperature.

- The emissions in THC increase when the concentration of VOC in air increases. Nevertheless, high VOC concentrations lead to high DRE (for a given temperature).
- The emissions of CO increase when the VOC concentration in air is increased, even when a constant CC output temperature is maintained. This increase is approximately linear.
- The emissions of NO decrease when the concentration of VOC in the air flux is increased; this should be a good reason to practise VOC pre-concentration before treatment by a RI.

In an application where the aim is to obtain a DRE of 99.4, such as the one studied here, the CC temperature has to be kept high because of CO emissions, and not because of unburned HC emissions.

From a series of experiments in which no VOC was injected, it has been shown that NO_x are formed by the burner alone, and not by the combustion of VOCs. The burner does not generate either CO or unburned hydrocarbons.

The present results indicate that the turbulence level inside the CC plays an important role in both the oxidation of HC into CO or CO₂, and in the oxidation of CO into CO₂.

We think that while the absolute values will differ, the tendencies of the present results can be generalised to VOCs other than ethyl acetate. Indeed, the VOC are known to rapidly crack into lighter HC species, which are rapidly oxidised into CO. As we have shown here, it is the oxidation of CO into CO₂ that is difficult to achieve, while high destruction efficiency for hydrocarbons is easy. This last reaction will occur in the same way regardless of the type of VOC that is burned.

References

- [1] E.N. Ruddy, L.A. Carroll, Select the best VOC control strategy, *Chemical Engineering Progress* 89 (7) (1993) 28–35.
- [2] D.R. Van Der Vaart, E.G. Marchand, A. Bagely-Pride, Thermal and catalytic incinerators for the controls of volatile organic compounds, *Journal of Air and Waste Management Association* 41 (1) (1991) 92–98.
- [3] L. Bussac, E. Caverne, Veille technologique sur le traitement des COV, CERUG/GDF 962696, 1996.
- [4] Y. Kara, Oxydation thermique des Composés Organiques Volatils en épurateur récupératif—Optimisation des outils de dimensionnement, Thèse de Doctorat (PhD) de l'INP Toulouse, France, 2003.
- [5] S. Salvador, Y. Kara, J.-M. Crussol, Improving VOC recuperative incinerators performances by increasing turbulence levels inside the combustion chamber—Experimental results, *Applied Thermal Engineering* 25 (2005) 1871–1881.
- [6] G.E.P. Box, W.G. Hunter, J.S. Hunter, *Statistics for experiments*, J. Wiley, New York, 1978.
- [7] G.E.P. Box, E. Drapper, *Empirical Model Building and Responses Surfaces*, J. Wiley, New York, 1987.
- [8] P. Dagnelie, A. Kblinski, D. Mathieu, R. Phan-Tan-Luu, J.P. Gauchy, G. Saporta, in: Fine J. Drosbecke, G. Saporta (Eds.), *Plans d'expériences: application à l'entreprise*, Technip, Paris, 1998.
- [9] P.W.M. John, *Statistical Design and Analysis of Experiments*, Macmillan, New York, 1971.
- [10] D.C. Montgomery, *Design and Analysis of Experiments*, J. Wiley, New York, 1996.
- [11] Y. Kara, S. Salvador, J.-M. Commandré, Thermal oxidation of volatile organic compounds in recuperative incinerator—optimisation of the operating parameters and of the geometry, *Congress Waste-Eng*, Ecole des Mines d'albi, mai 2005.
- [12] R. Phan-tan-Luu, D. Mathieu, M. Sergent, *Méthodologie de la recherche expérimentale*, LPRAI SARL, 1999.
- [13] S. Salvador, Y. Kara, J.-M. Commandré, Reduction of NO emissions of a VOC recuperative incinerator by dilution of the fuel supply, *Applied Thermal Engineering* 24 (2004) 245–254.
- [14] Arrêté relatif aux prélèvements et à la consommation d'eau ainsi qu'aux émissions de toute nature des installations classées pour la protection de l'environnement soumises à autorisation, Ministère de l'aménagement du territoire et de l'environnement, *Journal Officiel de la République Française*, arrêté du 2 février 1998, modifié le 29 mai 2000.
- [15] D.M. Stansel, N.M. Laurendeau, D.W. Senser, CO and NO_x emissions from controlled-air burner: experimental measurements and exhaust correlations, *Combustion Science and Technology* 104 (4–6) (1995) 207–234.