ODORS AND GAS EMISSIONS DURING THE CERAMIC SINTERING OF SEWAGE SLUDGE. GUIDELINES FOR INDUSTRIAL IMPLEMENTATION

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Abstract

Numerous research reported in the scientific literature show the feasibility of inerting sewage sludge in ceramic matrices as structural or red ceramic material for construction. However, its industrial implementation is hampered by a social rejection from the environmental movements.

Is this process an undercovered waste incineration or rather a pyrolysis process with reduced environmental impact? If there are countless incinerators, why does the manufacturing process of clay bricks with sewage sludgehave legislative difficulties and generates numerous social conflicts?

This study analyzes the data of gaseous, particulate matter and odors emissions produced in the sintering of clay/sewage sludge ceramic pieces. According to laboratory tests, VOC emissions for the clay/sludgematerial were in general higher than those from conventional ceramics. However, no VOC exceeded the threshold limit values and only a few compounds showed concentrations that exceeded their odor detection threshold. Besides, three inorganic pollutants exceeded the maximum levels (NO_x, suspended particles and HCl). In a test at industrial level, the most important emissions were those ofCO(1100 mgNm^{-3}), TOC(1085 mgNm^{-3}) andHCl(71 mgNm^{-3}).

Finally, from this experience, the main guidelines to be followedby any implementation of industrial ceramic plantsto be respectful with the environmentrules are proposed.

Keywords

Sewage sludge, clay bricks, sintering, odors, gas emissions, industrial production, guidelines.

Introduction

The huge amounts of sewage sludges from wastewater treatment plants (WWTP) pose a serious environmental challenge and entail high economic costs to society in relation to their treatment and disposal since they are toxic and hazardous wastes (Han et al., 1991) (Fig. 1).

Fig. 1 here

The valorisation of these sewage sludges as raw material for obtaining ceramic material for the construction has been extensively studied over the past two decades (Liew et al.,2004; Szoke and Muntean,2009; Lin and Weng,2001; Jordan et al.,2005). In all cases, the technical feasibility of producing these products for the construction obtained from wet mixtures of clays with added waste in a maximum percentage of 25 wt.% has been proved. Some properties of these ceramic products are the following

(Devant et al.,2011): a) acceptable resistance to compression according to standards (> 100 kpcm^{-2}),b) significant reduction in thermal conductivity over conventional ceramic (0.4 W m^{-1}K^{-1}, approximately), c) reduction of sound transmission coefficient, and d) low levels of leaching of heavy metals and other polluting compounds, below those specified in regulations (Gèric et al., 2012).

Gaseous emissions and odors are the main environmental concern in the production process of ceramic material with added sludge. Nevertheless, regarding this issue, the process of ceramization of sludge is more advantageous than other processes of "waste reduction" such as incineration, pyrolysis, gasification and wet oxidation (Fytili and Zabaniotou, 2008).In addition, with regard to CO_2 emissions, sewage sludge is considered biomass, and therefore their emissions are considered as “carbon neutral” when accounting for greenhouse gas (GHG) emissions and limits set in the Kyoto Protocol (United Nations, 2015).

It is often discussed whether the thermal process during ceramic firing responds to a process of incineration or pyrolysis. It is understood that the mixture of clay and sludge, once extruded, has a low oxygen content. Therefore,its thermal process would mostly correspond to a pryrolysis (Samolada and Zabaniotou, 2014). However, a longer residence time in the oven would destroy many of the complex compounds of organic nature in a tunnel kiln type Hoffman.

If there are countless incinerators installed in EU (Fig. 2), why does the manufacturing process of clay bricks with sewage sludge find legislative and social difficulties?

Fig. 2 here

The objectives of the present work are related to the analysis of the problem of the gaseous and odorous emissions as a tool for obtaining design guides to be used in the planning of future plants (or adaptation of the existing ones)of ceramic productswith added sewage sludge for building to be used both inenvelopes and structures (clay bricks, red ceramics, etc.). It is important to point out the need to make preliminary studies in laboratory because the gaseous emissions and odors are related to the proportions of eachraw materialin each particular case.

Materials and methods

A set of ceramic pieces were produced in order to carry out several tests. Crushed clays, urban sewage sludge (from biological treatment) and forest debris, with ratios of 80:15:5 in dry weight, respectively, were mixed with water sprayed in fine droplets in a 10-liter mixing bowl (Controls, model BT72). This mixture, that will be called “clay/sludge”, was moistened until optimal humidity was reached to get a homogenous material. Then, it was extruded under high-pressure (10 atm) (extruder Verdés, model 050-C) obtaining rectangular bars that were cut in test pieces of 5 or 12 cm long. The test pieces were dried at ambient air temperature (laboratory conditions) during 24 hours and then in a stove (Raypa, model DO-40) at 100°C during 24 additional hours. Finally, they were fired in a propane oven (Formagas, model HG-150) at a heating rate of 160°C h^{-1} from ambient temperature to 1000°C. Test pieces were inside the oven for 3 hours at 1000°C, and then a further 12 hours until they had cooled down to ambient temperature.

Gases produced during the firing step were sampled and analyzed following the U.S. EPA Reference Methods (EPA, 1994). Gas samples were taken from the oven stack. Stack diameter was 300 mm. Sampling hole was located at 6.9 diameters from the lower disturbance and 3.1 diameters from the upper disturbance. Three samples were obtained during firing one for each temperature range: 20-500°C, 500-940°C and 940°C. Besides, three more samples were taken for a conventional ceramics (100% clay) that will be called “clay”, one for each of those temperature ranges. All samples were analyzed using automated thermal desorption coupled to gas chromatography with mass selective detection. A detailed list of sampling and analytical equipments, as well as the methods followed for sampling and analysis can be found in Cusidó et al. (2003).

Results and discusion

Volatile organic compound (VOC) and inorganic compound emissions were determined during the bench firing process for both, clay/sludge and the regular clay pieces (see Table 1). As expected, VOC emissions from the clay/sludgematerial were in general higher than those from conventional ceramics. However, no VOC exceeded the threshold limit values with time weighted average (TLV-TWA). Only a few compounds (methyl mercaptane, dimethyl disulfide, acetic acid) had concentrations that exceeded their odor detection threshold (OD). In the case of the clay-brick firing, no VOC concentration higher than its OD or odor recognition threshold (OR)was detected, and obviously the values obtained were due to the propane gas used during furnace process. Some differences can be observed related to those obtained by direct sewage sludge burned at one fluidized-bed incinerator, especially in more complex compounds as benzene, toluene, ethylbenzene, acrylonitrile and acetonitrile (Tirey et al., 1991).

Table 1 here

As to the inorganic compound emissions, Table 2 shows the mean values of emission concentration obtained for the major compounds during the firing of both ceramics. For comparison purposes, their maximum limit values allowed by the most severe present legislation in Catalonia (solid waste incineration) are also indicated (DOGC, 1994).Inorganic emissions from the clay/sludgematerial were again higher than those from the clay-brick firing, except for the hydrogen fluoride (HF), which was therefore more due to the clay than to the wastes introduced in the clay/sludge mixture.

Table 2 here

Only three pollutants exceeded the maximum levels (Table 2): NO_x, suspended particles and HCl. It is well known that NO_x emissions are extremely sensible to the flame temperature and to the atmosphere in the oven (Patrick, 1994; Niessen, 1995). These variables were controlled by hand in the laboratory. However, since NO_x emissions can be better controlled in a industrial oven, it is expected that during industrial implementation values below the maximum limit allowed will be found. In the case of suspended particles, their concentration is highly dependent on the oven type (recirculations, internal streams, etc.), and therefore it is difficult to extrapolate results to a different scale. As to the high HCl concentration in emissions, its value is directly proportional to the percentage of sludge in the clay/sludge-brick. This problem is also present in the direct incineration of sludges (Wherther and Ogada, 1999), and it is mainly related to the use of chlorinated products as a correcting factor in the waste water treatment plants (although this practice is expected to be gradually reduced in the future).

The implementation of any production plant of clay/sludge brick must be accompanied by specific measures to minimize presence of contaminants in the air (emission) and the assessment of their potential area of impact in the surroundings (immission), which might affect the population directly or indirectly (through the food chain) in the medium and long term.

Since no specific regulations exist to be applied to plants leading with sewage sludge, the applicable regulations would be those regarding emission limits for urban waste incinerated promulgated by the EU (which are comparable to the limits established by the U.S. EPA for incinerators). See Table 3. Nevertheless, the process of ceramizationcould be assimilated to a thermal process of pyrolysis where emissions are significantly lower to those for an incineration.

Table 3 here

In relation to the exposed population three situations should be considered:

- Local population who can breathe emissions.

- Plant workers, especially those that perform maintenance tasks and control of installations.
- The regional population who may be affected by persistent contaminants and bioaccumulation of systemic pollutants (without adverse effects) and by the possible carcinogenic compounds.

Therefore it is always necessary to perform previous studies at the local level. Laboratory-scale studies of gaseous emissions and odors, emissions studies in plant and timed immission measurements according to existing regulations using as reference the corresponding ones to incinerators, by adapting them to the production levels of each plant (typically about 30 tonnes of sewage sludge per day in a small/medium plant).

In this regard, it can be mentioned an industrial scalelong-term studyconducted in a ceramic plant,CEASA(Papiol, Barcelona), which was producing bricks during 10 days from 240 t clay/sludge mixture with the same formulation than the oneabove mentioned used in laboratory (Devant, 2003) (Figs. 3 and 4). Its emissions containedhigh levels ofCO(1100 mgNm^{-3}),TOC(1085 mgNm^{-3}) andHCl(71 mgNm^{-3}). These levels are aboveofthe regulated emission limits. It was detected the existence ofunburnt organic matterwhich was responsible for strong smells.HClemissionwas associated tothe presence of chlorinein both,claysand sewagesludge,which can be correctedby adapting thefurnacefiring curveandfilters.From the analysis of the emitted gases, a total of 57 VOC were found. The most significant compounds were: benzene (C_6H_6, 1172 mgNm^{-3}), dimethyl disulfide ($C_2H_6S_2$, 1383 mgNm^{-3}), toluene (C_7H_8, 926 mgNm^{-3}), 4-methylpentanenitrile ($C_6H_{11}N$, 697 mgNm^{-3}), benzonitrile + isociane-benzene (C_7H_5N + C_7H_5, 1003 mgNm^{-3}).

Fig. 3 here

Fig. 4 here

For the study of the dispersion of gases, particles, and odors mathematical models for continuous point sources can be applied, being the Gaussian plume modela well-known option (Stockie, 2011). There isa number of available computer programs based on this model, such as some of the recommended ones for the Support Center for Regulatory Atmospheric Modeling of the U.S. EPA, like for instance AERMOD or CALPUFF (SCRAM, 2015). More sophisticated models (mesoscale 3D models) solve the momentum equations for horizontal wind components, the incompressible continuity equation for the vertical velocity in complex terrains, and scalar equations for potential virtual temperature, specific humidity of water vapors, cloud water and rain water, but require more computing power than the previous ones. An example of this type of modeling with TAPM (The Air Pollution Model) (Hurley, 2005) is shown in Fig. 5.

Fig. 5 here

The previous local study should follow these recommendations:

i) On the workplace (regarding effects on plant personnel): avoiding contact during the procedures including waste handling; burndown/cooldown; wearing masks and gloves to prevent viral infections; using a tower for biological washing of gases that are introduced into the kiln, cyclones to capture particulate emissions from stacks and perform a post-combustion cycle minimum of 3 minutes between 800 and 900°C to destroy the VOC, dioxins and furans and oxidize CO.A system of gas/gas exchanger is also recommended for the utilization of gases for drying or pre-baking (eg, Solvay NEUTREC® system).Finally, havingbiological filters or othersfor removing odors(if the site is close to inhabited areas), and apply all other considerations regarding the treatment of toxic and hazardous wastes as described by EPA (1990) and subsequent regulations.

ii) Regarding the location of the plant (either new or adapted from a pre-existing one). Recommended locations would be in rural and sparsely populated environments; knowledge of the climatic characteristics of the site (solar radiation, humidity and, in particular, the existence of weather situations of temperature inversion of great importance in the selection of the location); thorough knowledge of winds. Wind causes a horizontal dispersion of air pollutants and determines the area that will be exposed to immission. Generally, the higher the wind speed,the lower the concentrations of pollutants at ground level since there would be a greater dispersion and mixing. However adverse weather conditions (closed wind circulations,such as sea breezes, or temperature inversions) can create negative conditions by increasing pollutant concentrations in certain areas due to convective vertical transport.

iii) Control of emissions/immissions. Monitoring plans should be established from the estimated or known emissions data (concentrations, flowrates, temperatures). The theoretical study will determine the main impacted areas that are usually found in the vicinity of 10 km at most. For the monitoring of the impacted area is desirable to establish sensors of the following parameters: a) continuous emission monitoring (CEM) of particles, SO_2, CO, O_2, NO_x, HCl among others depending on each case, b) on a regular basis or even exceptionally in our case, stack tests of dioxins, furans, heavy metals, being unnecessary measurements of Hg, c) environmental monitoring (air, water, soil, food, etc.) around the industrial plant in a random manner. Finally, it is recommended to facilitate inspections, certifications of qualification of the personnel as well as sanitary control, safety equipment, etc.

iv) Information policy.It is noticeable the existence of a strong social opposition to the implementation of the process of sludge inertization inceramic matrices. It often creates social alarm due to unfamiliarity and lack of information about this novel process. Establishing a line of serious and truthful information can help achieve the acquiescence of the surrounding population, in particular if they are thoroughly informed about the implementation of measures of environmental prevention and monitoringin order tominimize/reducerisk to the health of the inhabitants. On the other hand, to inform populationon the environmental benefits of the technology (reduction of GHG and waste in landfills) and the products obtained should contribute to the acceptance of the definitive industrial implementation of the new ceramic product.

Conclusions

The production of ceramic material by inertizing sewage sludge is a solution for eliminating pathogens, vitrifying heavy metals into a ceramic matrix and removing most part of VOC in the thermal sintering process. With its industrial implementation, a double goal can be achieved: final and secure disposal of a hazardous waste and its valorization in a commercial product of great demand.

The production of ceramic material for construction from clay/sewage sludge mixtures can be a real solution for the safe disposal of waste from WWTP. However, environmental issues seem to be an obstacle to its widespread implementation, despite its several advantages(economic, as a final product and from the environmental point of view).

Compared to the simple incineration of sludge (among other thermal processes associated) of frequent application and with higher pollutant loads, it is necessary to claim the option of valorisation of sludge as raw material for obtaining ceramic products of high production (such as clay bricks).

For the implementation of the industrial production, it is necessary to carry out laboratory studies, since these residues (as well as the clays) have very varying characteristics depending of their site of origin. The application of the regulations of each country, as well as the existence of technologies for the treatment of gases, odors and particulates, would allow a clean production in accordance with legal standards. In no case it will be permissible to put in risk the users of these products, neither to affect the population living around the location of the plant.

The follow-up to the guidelines that have been proposed should be sufficient to ensure the industrial operation of a new industrial production line, whose pollutant load can be considered lower than that of any other currently available process of thermal treatment of residues. In addition, its production implies a complete elimination of sludge and its valorization into a commercial product with added value.

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Table 1. VOC emitted during the firing process [a].

Family	VOC	Formula	Clay/sludge-brick firing emissions (μg m^{-3})	Clay-brick firing emissions (μg m^{-3})	TLV-TWA (μg m^{-3})	OD [b] (μg m^{-3})	OR [c] (μg m^{-3})
Chlorinated hydrocarbons	Trichlorofluoromethane	CCl_3F	0.0	571.9	5620		
	Chloromethane	CH_3Cl	536.5	0.0		20462	20462
	Dichloromethane	CH_2Cl_2	1846.7	0.0	174000	550008	790637
	Trichloromethane	$CHCl_3$	179.5	940.5			
Mercaptans	Methylmercaptan	CH_3SH	16.0	0.0	980	1.1	2.0
Sulfides	Carbon disulfide	CS_2	728.8	142.7	31000	1306.3	1306.3
	Dimethyl disulfide	$S_2(CH_3)_2$	68.4	0.0		7.7	
	Dimethyl trisulfide	$S_3(CH_3)_2$	0.0	0.0		5.2	
Thiocianates	Methyl thiocyanate	CH_3SCN	169.9	0.0			
Aliphatic ketones	Propanone	C_3H_6O	1142.4	0.0		147160	308561
	3-Methyl-3-buten-2-one	C_5H_8O	151.8	0.0			
Aliphatic aldehydes	2-Methyl propanal	C_4H_8O	233.0	0.0			
	2-Methyl propenal	C_4H_6O	303.5	0.0			
	3-Methyl butanal	$C_5H_{10}O$	305.6	0.0			
	Hexanal	$C_6H_{12}O$	104.7	0.0			
	Heptanal	$C_7H_{14}O$	183.8	0.0			
Aromatic aldehydes	Furfural	$C_5H_4O_2$	255.4	0.0	7900	2498.6	2498.6
	Benzoaldehyde	C_7H_6O	594.2	0.0			
	Hydroxybenzaldehyde	$C_7H_6O_2$	5.3	0.0			
Aliphatic nitriles	Acetonitrile	C_2H_3N	1688.5	0.0	67000		

	Benzonitrile	C_7H_5N	318.5	0.0			
Aliphatic acids	Acetic	$C_2H_4O_2$	2533.8	0.0	25000	181.7	
	2-Methylpropanoic	$C_4H_8O_2$	32.1	0.0			
Aliphatic esters	Methyl acetate	C_3H_6O	238.3	0.0	606000	427238	
Aromatic amines	Pyrazine	$C_4H_4N_2$	160.3	0.0	16000	2133.7	2392.4
	Pyridine	C_5H_5N	454.2	0.0			
	4-Methylpyrazine	$C_5H_6N_2$	226.6	0.0			
	4-Methylpiridine	C_6H_7N	56.6	0.0			
Aliphatic amides	Acetamide	C_2H_5NO	237.2	0.0			
	3-Methylbutanamide	$C_5H_{11}NO$	0.0	0.0			
Monoterpenes	α-Pinene	$C_{10}H_{16}$	11.8	24.5		64	
Linear aliphatic hydrocarbons	N-nonane	C_9H_{20}	160.3	0.0	1050000		
	N-decane	$C_{10}H_{22}$	536.5	37.3			
	N-undecane	$C_{11}H_{24}$	0.0	37.3			
	N-dodecane	$C_{12}H_{26}$	89.8	0.0			
	N-tridecane	$C_{13}H_{28}$	361.2	0.0			
	N-tetradecane	$C_{14}H_{30}$	0.0	0.0			
	N-pentadecane	$C_{15}H_{32}$	0.0	0.0			
	N-hexadecane	$C_{16}H_{34}$	245.8	0.0			
Branched aliphatic hydrocarbons	2,2-Dimethylpentane	C_7H_{16}	5686.4	10693.3			
	Nonanes	C_9H_{20}	0.0	0.0			
	Decanes	$C_{10}H_{22}$	0.0	0.0			
	Undecanes	$C_{11}H_{24}$	0.0	0.0			
Polycyclic hydrocarbons	Decahydronaphthalene	$C_{12}H_{18}$	0.0	0.0			
	Methyl-decahydronaph-	$C_{13}H_{20}$	0.0	0.0			

	thalene						
Monocyclic aromatic hydrocarbons	Benzene	C_6H_6	961.8	92.7	32000	194712	
	Toluene	C_7H_8	582.4	182.1	188000	6023.9	41414
	Ethylbenzene	C_8H_{10}	56.6	0.0	434000	2602.7	2602.7
	m+p –Xylene	C_8H_{10}	190.2	22.4	434000	86757.2	
	Styrene	C_8H_{10}	166.7	0.0	434000	85120.3	

[a] Units are referred to standard conditions: T = 25ºC and P = 1 atm.

[b] Odour Detection Threshold.

[c] Odour Recognition Threshold.

Table 2. Mean emission levels of selected major inorganic compounds during the firing process and maximum limits allowed in special wastes incinerators in Catalonia [a].

Pollutant	Units	Clay/sludge brick	Clay brick	Limit allowed [b]
Particles	mg Nm^{-3}	48.3	11.4	20
SO_2	mg Nm^{-3}	43	8.4	200
CO	mg Nm^{-3}	83	26	125
NO_x	mg Nm^{-3} NO_2	811	805	616
HCl	mg Nm^{-3}	112	0.7	60
HF	mg Nm^{-3}	1.2	1.6	4
Cd+Tl	mg Nm^{-3}	0.007	n.d.[c]	0.1
Sb+As+Pb+Cr+Co+ +Cu+Mn+Ni+V+Sn	mg Nm^{-3}	0.44	0.02	1

[a] Units are referred to normal conditions: T = 0ºC, P = 1 atm, 11 v.% O_2 and dry gas.

[b] Decree 323/1994 of the Generalitat de Catalunya that regulates the facilities for waste incineration and determines their atmospheric emission limits. Values correspond to the strictest limits, i.e., those applicable to special waste incinerators (DOGC, 1994).

[c] “not detected”.

Table 3. Regulatory limits for pollutant emissions from incinerators (Batterman, 2004)

Pollutant	Units	EPA limits			EU limits		
		Small	Medium	Large	Daily	Hourly	4 hour
Particulate matter	mg dscm^{-1}	69	34	34	5	10	
Carbon monoxide	ppm(v)	40	40	40	50	100	
Dioxins/Furans	ng dscm^{-1} total	125	25	25			
	ng dscm^{-1} total TEQ	2.3	0.6	0.6			0.1
Organics					5	10	
					total Cl		
Hydrogen chloride	ppm(v)	15	15	15	5	10	
	% reduction	99%	99%	99%			
Sulfur dioxide	ppm(v)	55	55	55	25	50	
Nitrogen oxides	ppm(v)	250	250	250	100	200	
Lead	mg dscm^{-1}	1.2	0.07	0.07			
	% reduction	70%	98%	98%			
Cadmium	mg dscm^{-1}	0.16	0.04	0.04			0.05
	% reduction	65%	90%	90%			
Mercury	mg dscm^{-1}	0.55	0.55	0.55			0.05
	% reduction	85%	85%	85%			

Notes:

- U.S. EPA capacities: small < 91 kg h^{-1}; medium = 91 – 227 kg h^{-1}; large > 227 kg h^{-1}

- dscm = dry standard cubic meter; ppm(v) = parts per million by volume; TEQ = toxic equivalent, concentrations at 7 v.% O_2.

Fig. 1. Sewage sludge from WWTPwhich daily production is equivalent to 1 kg per inhabitant. It is a hazardous waste.

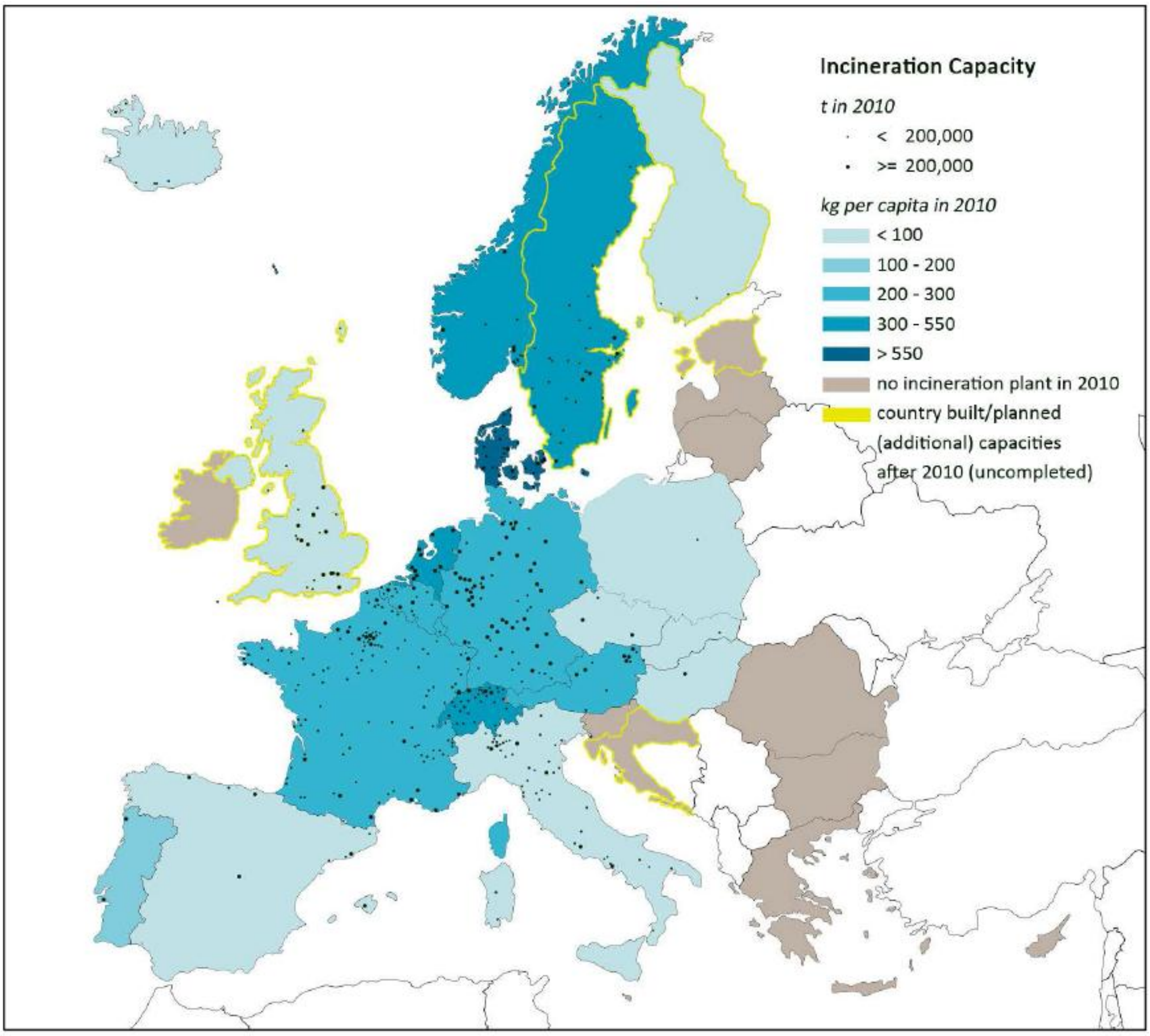

Fig. 2. Municipal solid waste (MSW) incineration capacity per capita and country in Europe and MSW generation in 2010 (Reichel et al., 2014).

Fig. 3. Clay bricks made from biological treatment sludge and shreddedforest residues (sawdust) in a ceramic industrial plant (Devant, 2003).

Fig. 4. Detail image of the texture of the clay/sludge material after sintering, showing great porosity due to the organic matter present in initial composition. This gives interesting thermal and acoustic properties to the final product (Cusidó et al., 2003).

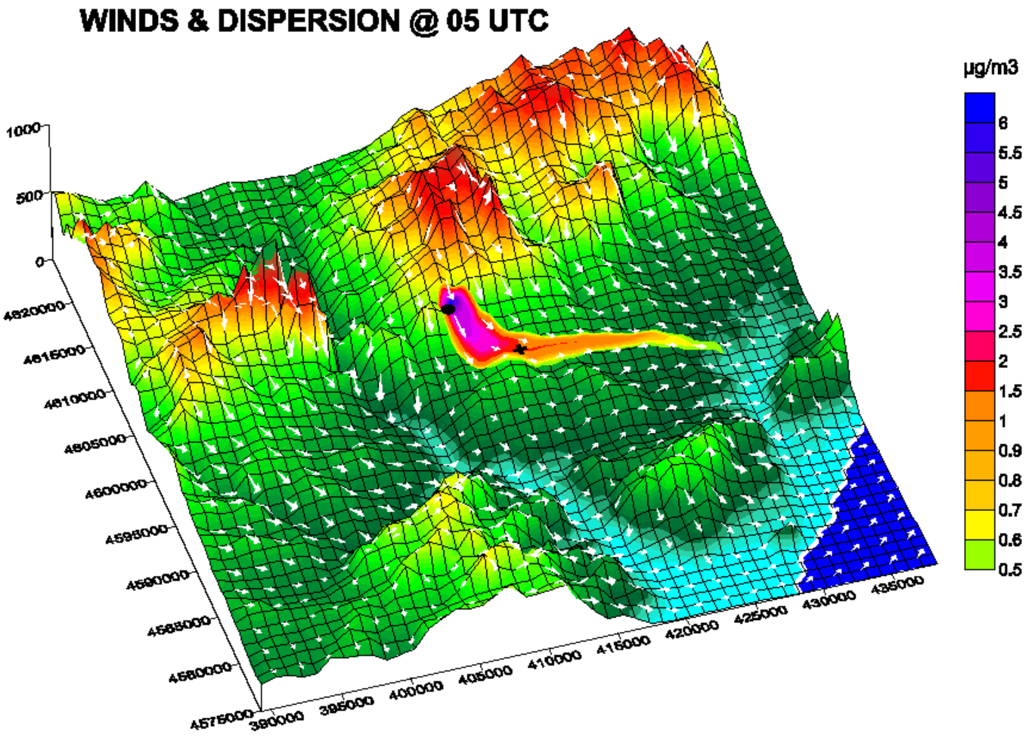

Fig. 5.A dispersion simulation of the plume associated to the emissions of an odorous source in the Barcelona area using TAPM atmospheric dispersion model (Gallego et al., 2008). It shows how the atmospheric conditions play a key role regarding the dispersion of the emissions, together with the surrounding orography. Nighttime drainage and katabatic winds originated in the slopes of the neighboring mountains disperse the plume downhill, which may result in annoyance to the surrounding population. During daytime hours, the penetration of the sea breeze and the formation of upslope winds during daytime, together with the deepening of the mixing layer, will move the plume towards the north of the domain and dilute odor concentrations and neighbor annoyance.