Extension of the Project for Obtaining Bioethanol from Citrus Waste

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ABSTRACT

The production of citrus fruit in Spain generates more than 0.5 million tons of waste. This is mainly due to rejects when packing fresh fruit and the waste generated by processing industries. That is the reason why an important challenge for the citrus sector is the use of this waste to unlock its economic value and eliminate the environmental problems that it may generate. The Instituto de Ingeniería de Alimentos para el Desarrollo (IuIAD) at the Universitat Politècnica de Valencia (UPV) has developed a system for transforming citrus waste into bioethanol and high value-added products such as essential oils. The purpose of this study was to find the most appropriate sites to implement a bioethanol industry from citrus waste. A spreadsheet was developed from the industrial design for transforming citrus waste into bioethanol. This spreadsheet allowed us to calculate the bioethanol mass flow from a known oranges, tangerines, lemons and limes and grapefruits flow waste. Total waste of the main citrus producers was estimated as the sum of the rejects when packing fresh fruits and the wastes generated by processing industries. Total waste value (oranges, tangerines, lemons and limes and grapefruits) was multiplied by the ratio between mass flow and citrus waste flow in order to calculate the bioethanol production in each country. The results suggest that Brazil, USA, Mexico, China, India and Italy would be the most appropriate countries to implement a bioethanol industry from citrus waste.

Keywords: citrus, citrus waste, bioethanol, by-products, industrial citrus process

INTRODUCTION

The use of petroleum-based sources of energy causes both environmental and economic concerns. Energy consumption has increased considerably in recent years as the world population grows and more countries have become industrialized (Sun and Cheng, 2002). This, together with the stagnation of fossil fuel reserves, has led to rising oil prices. In addition, fossil fuels generate greenhouse gas emissions which are responsible for the climate change. As a result, the search into renewable, sustainable and environmentally-friendly energy sources such as biomass has been encouraged. According to several authors, transportation fuels such as bioethanol or biodiesel seem to be the most suitable alternative in the short-term (Alvira et al., 2010; Sun and Cheng, 2002).

Bioethanol is a renewable fuel that can be obtained by fermenting sugar-rich, amylaceous or lignocellulosic biomass of different origin (Gonzalez-Garcia et al., 2009; Mussatto et al. 2010) However, sustainability of current production of bioethanol has become controversial due to the fact that it mostly relies on energy crops such as corn or sugarcane. This debate has caused an increasing interest in lignocellulosic biomass as an alternative for bioethanol production since it does not compete with food and it is usually less expensive than other agricultural feedstock (Alvira et al., 2010). Agricultural, industrial or forestry wastes are in this group and appear as a potential source for low-cost bioethanol production (Sun and Cheng, 2002).

In particular, up to 1,28x10^8 tons of citrus were produced worldwide in 2011 (FAOSTAT, 2011) and around 50% of the raw material becomes waste consisting of discarded fruits, peels, seeds and segments membranes (Braddock, 1999). This is mainly due to rejects when packing fresh fruits and the waste generated by processing industries. This waste contains high concentrations of biodegradable and suspends solids which cause a serious pollution problem if they are not disposed of properly because of their high Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD). However, this lignocellulosic material contains polysaccharides such as cellulose, hemicellulose and pectin, as well as several mono and disaccharides, the main ones being glucose, sucrose and fructose which are easily-fermentable by *Saccharomyces cerevisiae* to produce ethanol (Wilkins et al., 2007). The Institute of Food Engineering for the Development at the Polytechnic University of Valencia (Spain) has developed a system for transforming citrus waste into bioethanol, purified water, cattle feed and high value-added products such as essential oils.

The aim of this study was to find the most appropriate sites to implement a bioethanol industry from citrus waste. For this purpose, the present study was focused on the system developed by the Institute of Food Engineering for the Development and specifically on the biofuel production line.

MATERIALS AND METHODS

Developing a calculation basis for the industrial design

First a process diagram based on the citrus by-product industrial plant designed by the IuIAD was drawn up. Then a calculation basis for oranges, mandarins, limes and lemons, and grapefruits was created considering the material and energy balances of each stage of the process.

Calculation of ratios

In order to calculate the bioethanol production ratios for each type of citrus fruit, the quotient between bioethanol mass flow output and citrus by-product inflow to the plant was calculated. Both values were obtained from the calculation basis for each citrus fruit.

Calculation of total waste from major citrus-producing countries

The waste generated by oranges, mandarins, lemons and limes and grapefruits was calculated for each of the world's twenty major citrus-producing countries. To that end we established that the waste generated by each of type citrus fruit was equal to the sum of fresh produce withdrawals and industry by-products.

Fresh produce withdrawals are due to the existence of citrus fruits that have sustained wastage due to plant diseases, pathologies or other reasons or which do not meet minimum quality standards for marketing. Since all citrus fruits behave in the same way, i.e. have similar respiration rates and therefore the same wastage percentages, Alfonso et al. (2010) set a ratio of 0.02 of citrus fruit withdrawn to citrus fruit produced based on a range of literature sources and checking with the industry (MARM, 2008; MARM, 2006).

To calculate the by-product generated by the industry, the amount of citrus fruit used for fresh packing was determined for each country and product. This quantity was multiplied by 0.21 for oranges, by 0.12 for mandarins and by 0.11 for the rest of citrus fruits based on Alfonso et al. (2010) criterion.

Calculating the potential volume of bioethanol for each citrus-producing country

This involved multiplying the waste mass of each citrus product by the ratio between the bioethanol outflow and the inflow of each by-product to the plant obtained from the calculation basis. The total volume of bioethanol for each citrus-producer country was obtained by adding up the total volume of bioethanol for each type of citrus fruit. Finally the countries were sorted in descending order in terms of the amount of bioethanol that could be produced.

RESULTS AND DISCUSSION

Developing a calculation basis for the industrial design

To draw up the industrial design diagram for the plant in Figure 1, both the line for the production of bioethanol from the liquid phase of the pressed citrus waste and the line for producing animal feed from the press cake were considered. Firstly, the citrus waste was mixed in a hammer mill with 0.6% (w/w) of $Ca(OH)_2$ to enable double pressing with a hammer press. The resulting solid phase was dried to obtain pellets while the liquid phases from the first and second pressing were mixed in a third tank and water, nutrients, sulphuric acid, steam and *S. cerevisiae* were added. The final mixture fermented for 50 hours at 30°C. The fermented product was diverted into a first and a second distiller at atmospheric pressure using indirect steam from which bioethanol at 80% (v/v) and vinasse and bioethanol at 96% (v/v) and water were obtained respectively. The vinasse was concentrated to 45° Brix in a WHE (Waste Heat Evaporator). The concentrate was collected in a storage tank from where it was pumped to the cake before the first and second pressing to make them easier.

[Position of fig 1]

To calculate the mass flow rate of bioethanol at 96% (v/v) from each citrus waste, the concurrent design of all the equipment was integrated in a single calculation basis considering the material and energy balances found at each stage. In addition the industrial plant was designed to process 35 tonnes of citrus waste per hour. Finally, the calculation bases were developed considering all these premises and the physicochemical characteristics of each waste, i.e. its fermentable and non-fermentable soluble solids, insoluble solids and water content. Figure 2 shows the calculation basis of orange waste in which known flow and composition data were entered in the shaded boxes. The uncoloured boxes were for the composition and mass flow rate data of each of the products in the process stages obtained from the material and energy balances.

[Position of table 1]

Calculation of ratios

Table 2 summarized the ratios between the bioethanol mass flow rate obtained for each of the citrus wastes using the calculation basis and the waste input mass flow rate to the plant.

[Position of table 2]

Calculation of total waste from major citrus-producing countries

Table 3 summarized the tonnes of oranges produced and by-products generated by the industry together with fruit wastage for the 20 major orange-producer countries in 2011 (FAOSTAT, 2011). It was found that the amount of available waste is very high in countries such as Brazil and USA, which both produce large quantities and have major juice industries (USDA, 2010). Other countries such as India, Mexico and Italy also had higher available waste than Spain, which in the case of Italy was also due to its large processing industry (ISTAT, 2010; MARM, 2010; USDA, 2010). Finally other countries with less available waste than Spain such as China, Indonesia and South Africa also topped 100.000 tonnes per year.

[Position of table 3]

The production and processing values for the major mandarin producer countries in 2010 (FAOSTAT, 2011) were shown in table 4. It was found that, in general, these figures are lower than those for oranges (FAOSTAT, 2011) and hence there is less available waste. Furthermore only China had considerably more available waste than Spain due its higher production. Behind Spain with lower waste figures were Mexico, Brazil, Japan, Turkey and Egypt, all with more than 20.000 tonnes of mandarin waste.

[Position of table 4]

Table 5 summarized the production (FAOSTAT, 2011), processing and total amount of waste values for lemons and limes in 2011. Here Argentina, Mexico and the USA were significantly ahead due to their high production (FAOSTAT, 2011) and the size of their processing industries (USDA, 2010), while India also recorded high volumes due to its large production (FAOSTAT, 2011).

[Position of table 5]

Finally grapefruit production (FAOSTAST, 2011), processing and total waste values for 2011 were shown in table 6. In this case the USA was in the lead due to its major processing industry, followed by China, Mexico and South Africa (USDA, 2010).

[Position of table 6]

Calculating the potential volume of bioethanol for each citrus-producing country

Table 7 summarized total potential bioethanol production from each type of citrus fruit waste and the total. Both Brazil at more than 164.106 litres per year and the USA with over 87.106 litres per year far exceeded Spain's potential for producing bioethanol from citrus waste. Other countries such as Mexico, China and India, albeit with potential bioethanol figures lower than Spain's, might be potential candidates for setting up bioethanol production from citrus waste. Other countries such as Argentina, South Africa, Italy, Greece, Egypt, Indonesia, Turkey, Iran and Pakistan have less potential for producing bioethanol from citrus waste, but all of them exceeded 3.000 litres per year of bioethanol. Finally Peru, Syria, Cuba, Colombia and Thailand are countries whose potential bioethanol production from citrus waste was greater than 1.000 litres per year.

[Position of table 7]

CONCLUSIONS

Brazil and the USA would be the most appropriate countries to implement a bioethanol industry from citrus waste because of their big citrus production and their strong orange juice industry.

Mexico, China, India and Italy would be strong candidates to implement a bioethanol industry with an estimate production between $10x10^6$ and $17x10^6$ l/year. Most of the bioethanol production would be from oranges except in China because of the big processing industry cannery and juice and production of tangerines.

Argentina, South Africa, Greece, Egypt, Indonesia, Turkey, Iran and Pakistan would have an estimated lower bioethanol production between $3x10^6$ and $7x10^6$ l/year. Most of bioethanol production would be from oranges except in Aregentina because of her lemons and limes industry.

Peru, Syria, Cuba, Colombia and Thailand would be minor candidates to implement a bioethanol industry from citrus waste because of their lower production and processing industry.

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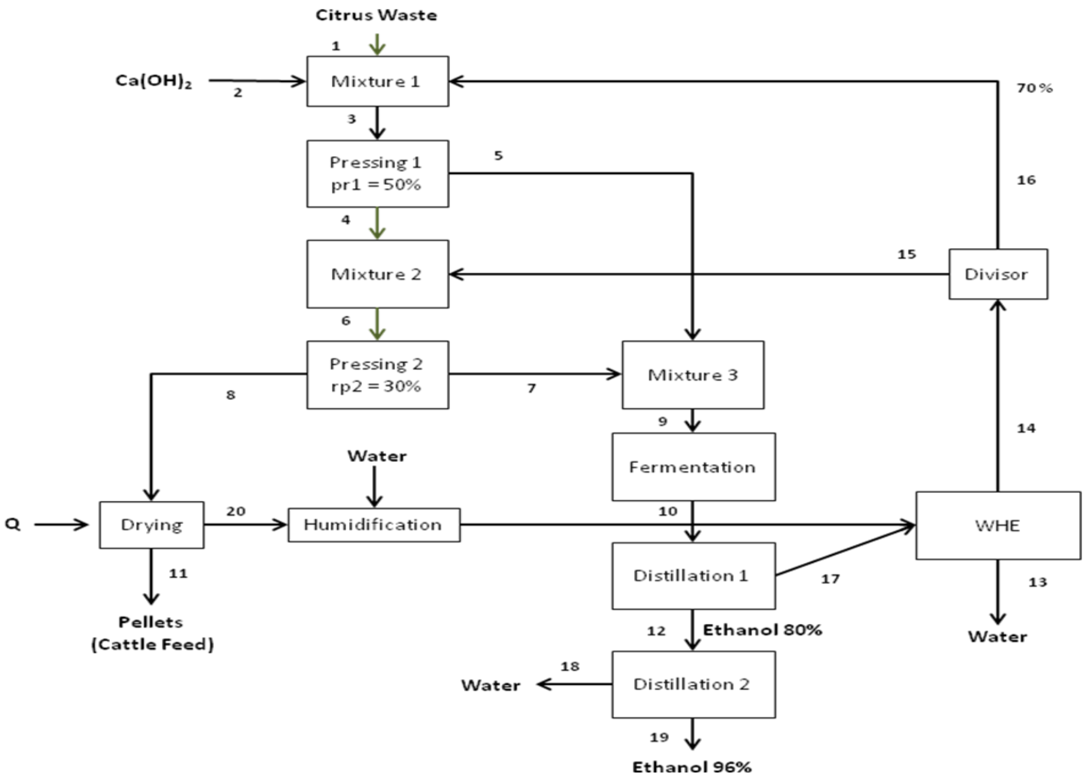

Figure 1. – Flow diagram of the industrial plant design for obtaining bioethanol from citrus waste.

Table 1. – Spreadsheet of the industrial plant design for obtaining bioethanol from orange waste.

Stream	Flow M (kg/h)	Water x_w	Ca(OH)2 x_{Ca}	Fermentable Soluble Solids x_{sf}	Non Fermentable Soluble Solids x_{sn}	Ethanol x_E	Insoluble solids x_{si}	SST Z_{ss} (Brix)
1	35.000	0,87	0	0,08176	0,0302	0	0,018	11,4
2	140	0,08	0,92	0	0	0	0	0,0
3	37.608	0,8461	0,0034	0,07609	0,0577	0	0,017	13,7
4	18.804	0,8286	0,0068	0,07452	0,0565	0	0,034	13,7
5	18.804	0,8635	0	0,07766	0,0589	0	0	13,7
6	19.862	0,8138	0,0065	0,07055	0,0774	0	0,032	15,4
7	5.958,5	0,8461	0	0,07336	0,0805	0	0	15,4
8	13.903	0,7999	0,0093	0,06935	0,0761	0	0,045	15,4
9	24.763	0,8593	0	0,07662	0,0641	0	0	14,1
10	23.886	0,8908	0	0	0,0664	0,043	0	6,9
11	3.090,5	0,1	0,0417	0,312	0,3425	0	0,204	86,7
12	1.276	0,2	0	0	0	0,8	0	0,0
13	19.084	1	0	0	0	0	0	0,0
14	3.525,9	0,55	0	0	0,45	0	0	45,0
15	1.057,8	0,55	0	0	0,45	0	0	45,0
16	2.468,1	0,55	0	0	0,45	0	0	45,0
17	22.610	0,9298	0	0	0,0702	0	0	7,0
18	212,67	1	0	0	0	0	0	0,0
19	1.063,3	0,04	0	0	0	0,96	0	0,0
20	10.813	1	0	0	0	0	0	0,0

Table 2. – Ratio between the bioethanol mass flow and the citrus waste mass flow.

$r_{bioethanol\text{-}orange\ waste}$ (w/w)	0,030
$r_{bioethanol\text{-}tangerine\ waste}$ (w/w)	0,029
$r_{bioethanol\text{-}\ lemon\ and\ lime\ waste}$ (w/w)	0,014
$r_{bioethanol\text{-}grapefruit\ waste}$ (w/w)	0,019

Table 3- Main orange producers and their total orange waste.

Rank	Country	Production (t)[1]	Industry (t)	Industrial waste (t)	Rejects (t)	Total waste(t)
1	Brazil	18.101.700	12.158.000[4]	3.890.560	362.034	4.252.594
2	United States	7.974.000	6.183.000[4]	1.978.560	159.480	2.138.040
3	India	6.268.100	658.151[4]	210.608	125.362	335.970
4	China	5.500.000	180.000[4]	57.600	110.000	167.600
5	Mexico	4.100.000	850.000[4]	272.000	82.000	354.000
6	Egypt	3.645.000	75.000[4]	24.000	72.900	96.900
7	Spain	2.669.355	500.342[3]	160.109	53.387	213.497
8	Italy	2.393.660	1.436.196[2]	459.583	47.873	507.456
9	Indonesia	2.032.670	213.430[4]	68.298	40.653	108.951
10	Turkey	1.710.000	100.000[4]	32.000	34.200	66.200
11	Pakistan	1.505.000	158.685[4]	50.779	30.100	80.879
12	Iran	1.502.820	158.455[4]	50.706	30.056	80.762
13	South Africa	1.414.590	243.133[4]	77.802	28.292	106.094
14	Morocco	849.197	6.252[4]	2.001	16.984	18.985
15	Argentina	833.486	189.519[4]	60.646	16.670	77.316
16	Greece	770.000	176.845[4]	56.590	15.400	71.990
17	Algeria	740.000	37.873[4]	12.119	14.800	26.919
18	Vietnam	729.400	0[4]	0	14.588	14.588
19	Syria	668.900	65.027[4]	20.809	13.378	34.187
20	Ghana	556.100	28.461[4]	9.107	11.122	20.229

Sources: [1]FAOSTAT, 2011; [2]ISTAT, 2010; [3]MARM, 2010; [4]USDA, 2010.

Table 4- Main tangerine producers and their total tangerine waste.

Rank	Country	Production (t)[1]	Industry (t)	Industrial waste (t)	Rejects (t)	Total waste(t)
1	China	10.142.430	420.000[4]	29.400	202.849	232.249
2	Spain	1.708.200	203.407[3]	14.238	34.164	48.402
3	Brazil	1.122.730	51.646[4]	3.615	22.455	26.070
4	Turkey	858.699	65.261[4]	4.568	17.174	21.742
5	Egypt	796.867	60.562[4]	4.239	15.937	20.177
6	Japan	786.000	94.000[4]	6.580	15.720	22.300
7	South Korea	614.871	85.000[4]	5.950	12.297	18.247
8	Pakistan	559.000	49.849[4]	3.489	11.180	14.669
9	United States	540.682	159.000[4]	11.130	10.814	21.944
10	Morocco	472.834	0[4]	0	9.457	9.457
11	Argentina	423.737	80.166[4]	5.612	8.475	14.086

12	**Mexico**	409.442	265.682(4)	18.598	8.189	26.787
13	**Thailand**	280.190	24.986(4)	1.749	5.604	7.353
14	**Iran**	276.138	24.625(4)	1.724	5.523	7.246
15	**Italy**	240.628	29.242(2)	2.047	4.813	6.859
16	**Peru**	221.324	10.128(4)	709	4.426	5.135
17	**Algeria**	185.800	0(4)	0	3.716	3.716
18	**Nepal**	174.867	0(4)	0	3.497	3.497
19	**Israel**	152.207	35.515(4)	2.486	3.044	5.530
20	**South Africa**	142.500	0(4)	0	2.850	2.850

Sources: (1)FAOSTAT, 2011; (2)ISTAT, 2010; (3)MARM, 2010; (4)USDA, 2010.

Table 5- Main lemon and lime producers and their lemon and limes total waste.

Rank	**Country**	**Production (t)(1)**	**Industry (t)**	**Industrial waste (t)**	**Rejects (t)**	**Total waste(t)**
1	**India**	3.098.900	153.819(4)	24.611	61.978	86.589
2	**Mexico**	1.880.000	305.000(4)	48.800	37.600	86.400
3	**Argentina**	1.300.000	973.000(4)	155.680	26.000	181.680
4	**China**	1.058.105	52.521(4)	8.403	21.162	29.565
5	**Brazil**	1.020.350	92.243(4)	14.759	20.407	35.166
6	**United States**	853.000	355.000(4)	56.800	17.060	73.860
7	**Turkey**	782.000	60.000(4)	9.600	15.640	25.240
8	**Iran**	706.800	35.083(4)	5.613	14.136	19.749
9	**Spain**	558.180	68.301(3)	10.928	11.164	22.092
10	**Italy**	522.377	73.133(2)	11.701	10.448	22.149
11	**Egypt**	318.111	0(4)	0	6.362	6.362
12	**Peru**	233.032	21.067(4)	3.371	4.661	8.031
13	**Sudan**	228.000	0(4)	0	4.560	4.560
14	**South Africa**	215.985	5.400(4)	864	4.320	5.184
15	**Thailand**	171.074	8.492(4)	1.359	3.421	4.780
16	**Chile**	155.000	14.013(4)	2.242	3.100	5.342
17	**Syria**	142.200	12.225(4)	1.956	2.844	4.800
18	**Lebanon**	113.100	9.724(4)	1.556	2.262	3.818
19	**Guatemala**	107.796	9.745(4)	1.559	2.156	3.715
20	**Colombia**	87.474	7.908(4)	1.265	1.749	3.015

Sources: (1)FAOSTAT, 2011; (2)ISTAT, 2010; (3)MARM, 2010; (4)USDA, 2010.

Table 6- Main grapefruit producers and their grapefruit total waste.

Rank	**Country**	**Production (t)(1)**	**Industry (t)**	**Industrial waste (t)**	**Rejects (t)**	**Total waste(t)**
1	**China**	2.600.000	0(4)	0	52.000	52.000
2	**United States**	1.090.000	520.000(4)	114.400	21.800	136.200
3	**Mexico**	430.000	100.000(4)	22.000	8.600	30.600
4	**South Africa**	370.000	167.000(4)	36.740	7.400	44.140
5	**Thailand**	294.949	0(4)	0	5.899	5.899
6	**India**	260.300	0(4)	0	5.206	5.206

7	**Israel**	225.000	104.000[4]	22.880	4.500	27.380
8	**Turkey**	210.000	2.376[4]	523	4.200	4.723
9	**Cuba**	137.660	121.967[4]	26.833	2.753	29.586
10	**Argentina**	130.000	62.000[4]	13.640	2.600	16.240
11	**Tunisia**	89.800	0[4]	0	1.796	1.796
12	**Brazil**	72.100	5.382[4]	1.184	1.442	2.626
13	**Bangladesh**	58.468	0[4]	0	1.169	1.169
14	**Iran**	46.500	0[4]	0	930	930
15	**Paraguay**	44.000	3.285[4]	723	880	1.603
16	**Spain**	43.200	2.512[3]	553	864	1.417
17	**Swaziland**	37.000	0[4]	0	740	740
18	**Belize**	36.000	2.688[4]	591	720	1.311
19	**Philippines**	33.472	0[4]	0	669	669
20	**Italy**	7.125	414[2]	91	143	234

Sources: [1]FAOSTAT, 2011; [2]ISTAT, 2010; [3]MARM, 2010; [4]USDA, 2010.

Table 7. World's main bioethanol producers from orange, tangerine, lemon and lime and grapefruit waste.

Rank	**Country**	**Total Bioethanol (l/year)**	**Bioethanol from orange (l/year)**	**Bioethanol from tangerine (l/year)**	**Bioethanol from lemon and lime (l/year)**	**Bioethanol from grapefruit (l/year)**
1	**Brazil**	164.331.569	162.718.262	952.000	598.780	62.527
2	**United States**	87.110.451	81.808.457	801.331	1.257.632	3.243.032
3	**Spain**	20.050.118	19.416.939	250.471	377.136	5.572
4	**China**	16.723.153	13.545.207	978.183	1.471.153	728.611
5	**Mexico**	16.635.553	6.412.928	8.481.053	503.410	1.238.162
6	**India**	14.453.650	12.855.319	0	1.474.371	123.959
7	**Italy**	10.346.504	8.169.099	1.767.499	376.166	33.740
8	**Argentina**	6.952.940	2.958.365	514.379	3.093.508	386.688
9	**South Africa**	5.302.858	4.059.506	104.074	88.269	1.051.009
10	**Egypt**	4.553.917	3.707.713	736.805	108.327	1.071
11	**Turkey**	4.168.824	4.168.824	0	0	0
12	**Iran**	3.869.210	2.533.030	793.954	429.767	112.458
13	**Pakistan**	3.713.238	3.090.220	264.603	336.271	22.144
14	**Greece**	3.671.282	3.094.697	535.669	40.916	0
15	**Indonesia**	2.994.931	2.754.575	135.332	54.283	50.741
16	**Peru**	1.508.042	1.308.107	34.509	81.731	83.695
17	**Cuba**	1.190.505	1.030.010	135.697	24.179	619
18	**Colombia**	1.094.225	202.222	201.939	38.124	651.940
19	**Syria**	1.079.909	726.429	345.342	7.662	476
20	**Thailand**	895.820	60.035	814.331	21.454	0