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Wet deposition and atmospheric mercury monitoring in Celestún, Yucatán, México, as part of the Global Mercury Observation System

Mercury concentration in ambient air - Results 2012

Fabrizio Sena, Gunther Umlauf, Martha Ramirez Islas, Juan Antonio Velasco Trejo, Flor Arcega Cabrera, Ismael Oceguera Vargas

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WET DEPOSITION AND ATMOSPHERIC MERCURY MONITORING IN CELESTÚN, YUCATAN, MEXICO, AS PART OF THE GLOBAL MERCURY OBSERVATION SYSTEM (GMOS)

MERCURY CONCENTRATION IN AMBIENT AIR – RESULTS 2012

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Summary

This report describes work conducted by the European Commission's Joint Research Centre in the contest of GMOS (Global Mercury Observation System). GMOS is an FP VII funded large-scale collaborative project aiming at the establishment of a Global Mercury Observation System including ocean-based, ground-based and atmospheric measurement activities under the umbrella of the GEO/GEOSS and the UNEP's Mercury program.

Within this 5 year project that started in 19 Nov 2010, JRC got the task to set-up a ground based station for measuring total atmospheric Mercury in Yucatán, Mexico. The system is fully automatic and transmits the acquired raw data via internet to the JRC for data analysis and evaluation prior to further reporting to the GMOS coordinator. Moreover, the time series obtained for Hg will be reported in the framework of Task Force on Hemispheric Transport of Air Pollution (LRTAP-Convention) co-chaired by the JRC (Climate change Unit).

In order to assist the JRC in the set up and routine maintenance of the Hg monitoring station, cooperation was initiated with the *National Institute of Ecology and Climate Change - Secretariat of Environment and Natural Resources (INECC-SEMARNAT)* in Mexico City and DUMAC foundation in Yucatán.

INECC agreed to assist the JRC in selecting an appropriate site partner, support maintaining the instrumentation, taking care of the wet deposition sampling/dispatch logistics, and supply meteorological data from the selected site.

Data regarding the total gaseous mercury concentration in air measured during the year 2012 at Celestún, Yucatán, México are here reported. The annual TGM average obtained from 44537 data was 1.047 ± 0.271 ng/m³. The minimum value (0.50 ng/m^3) was registered from February to May; the lowest monthly average (0.752 ng/m^3) was recorded in April. The maximum value (2.822 ng/m^3) was observed in March, while the highest monthly average TGM value was obtained in June (1.388 ng/m^3) .

1. Introduction

A sampling station for mercury monitoring in ambient air and precipitation was established as part of the Global Mercury Observation System (GMOS), following the project's requirement to include a remote background site in Latin America. The air data from January 28th to October 17th 2012 are reported in this document.

The National Institute of Ecology and Climate Change (INECC), as initial JRC project partner in Mexico, proposed the site of Celestún in the northeast of the Yucatán State at 20°51'33.33" N, 90° 23'33.51" E and an altitude of 3 m asl (Figure 1), just north of the border with the state of Campeche on the coast of the Gulf of Mexico.

The site is located next to a coastal lagoon and about 2000 m distance from the sea. Approximately at 1 km east of the site, and in the city of Merida (77.6 km NE) there are meteorological stations run by the National Meteorological Service. The Celestún site is characterized by a tropic climate with a dry season from March to May (< 50 mm) and a rainy season from June to October (>500 mm), and the 'Nortes' season from November until February characterized by moderate (20-60 mm) rainfall and intermittend high wind speeds > 80 km/h (Herrera-Silveira 1994). Typical average temperature means range from 20°C in January to 35°C in August with and avarage mean annual of 26.2°C (Herrera-Silveira 1994, 1996 cited in Young et al., 2005). Predominant winds during the day come from the East, by night, predominant winds come from the N-NE.

The site is located at the Celestún Biosphere Reserve, a 147,500 acre (600 km) land. This is a wetland that is the winter home for flamingos, herons and other bird species. The reserve contains a mixture of tropical savanna, low tropical sub deciduous forest and tular vegetation.

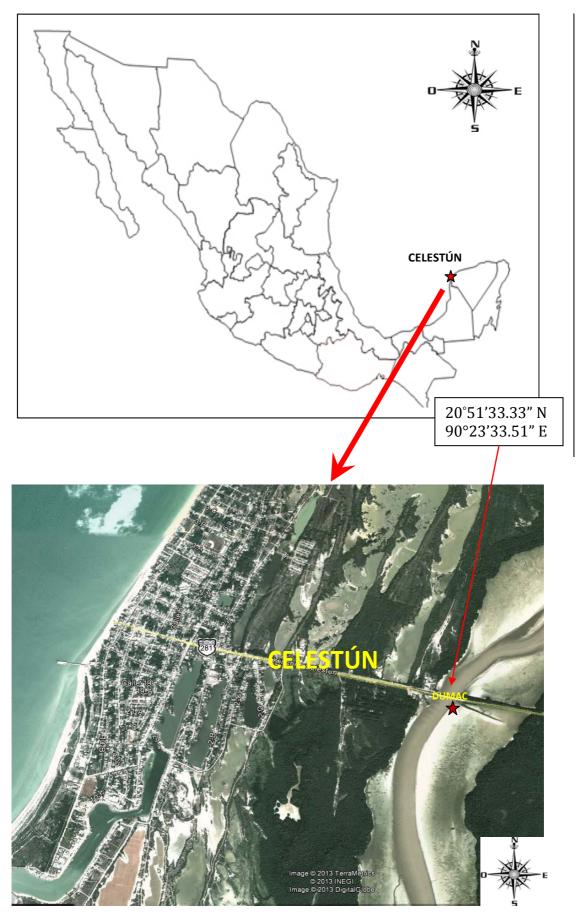


Figure 1, 2. Location of Ducks Unlimited de México A.C. (DUMAC) at Celestún, México

Celestún is a small town of approximately 6,000 habitants; the predominant economic activities are tourism, fishing and salt extraction. Nearby industrial activities are absent. The most important sources of mercury emissions in Yucatan are electrical energy generators (3 plants, 2 in Merida and 1 in Valladolid), and the cement industry at Merida. These mercury sources are more than 70 km away from the Celestún site. Forest fires are moderate in the state of Yucatán.

The selected location is at the "John E. Walker" Center for Research and Training on Natural Resources, owned by Ducks Unlimited de Mexico A.C. (DUMAC). This organization is a nonprofit association dedicated to the wetlands preservation for waterfowl (http://www.dumac.org/dumac/habitat/esp/index.htm). The DUMAC Center is located about 2 km away from the center of Celestún. The site is on the main road that leads to Mérida, the capital city of the state of Yucatán.

The DUMAC Center consists of a two-floor concrete structure; metal fences surround the area. The terrain is flat, with calcareous soil, scarcely covered by trees, bushes and grass (Figure 3).

At the roof, there is a measuring tower (Figure 4C) where the Hg sampling probe was installed at 10 m above ground. The air mercury analyzer TEKRAN 2537B was connected to the sampling probe trough a controlled heating tube. It was placed inside a shed with air conditioning (Figure 4A and 4B).



Figure 3. DUMAC Center

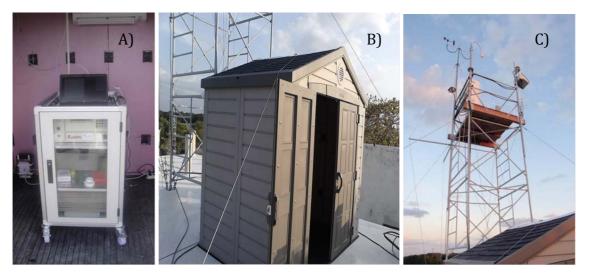


Figure 4. Equipment installed in DUMAC. A) Rack of the Tekran 2537B, B) Shed, C) Measuring tower.

The equipment installation for the ambient air measurements was done by the JRC from January 23 to 29 2012. A training course was given to the local partners from January 30 to 3 February 3rd. Measurements (continuous sampling at 5 min intervals) were initiated on February 4, 2012.

For a more detailed description of the site and the installation of the equipment please refer to the 2012 report entitled: Wet Deposition and Atmospheric Mercury Monitoring in Celestún, Yucatan, Mexico, as Part of the Global Mercury Observation System (GMOS), Equipment Location and Installation"

2. Methodology

Field measurements were done by JRC, INECC and DUMAC and UNAM joint technicians and experts, and took place in the Yucatán GMOS station during 2012. The Total Gaseous Mercury (TGM) measurements were carried out using a Tekran Model 2537B automatic mercury vapor analyzer, over a period of twelve months. Measurements and averages are summarized in Figure 5 and table 2.

The instrument was placed in an air conditioned shed (average temperature 26° C) and, it was connected to the outside probe by a thermal isolated tube, heated at 45°C. The outside probe sampled ambient air at the top of the measuring tower. The sampling line

was made PTFE tubing and its length was minimized, ensuring that no sharp bends were present.

In order to produce each individual measurement result, instrument sampled ambient air for a period of 5 min. The instrument makes the measurements by trapping TGM on absorption tubes followed by thermal desorption and detection by atomic fluorescence spectrometry. Instrument was installed, maintained and operated according to manufacturers' instructions and was calibrated and tested at the beginning of the measurement campaign and then every 71 hours thereafter.

Manual calibration tests were performed using a saturated mercury vapor calibration unit, which allows calibration of any low level gas phase measurement system. A thermoelectrically cooled reservoir, together with precision gas tight syringes, allows the injection of precisely known amounts of mercury (R. J. C. Brown et al. 2008).

TGM data were validated according to guidelines established by Global Monitoring System (GMOS). Weekly routine maintenance was conducted to ensure proper operation of the equipment throughout the study period.

Meteorological data (i.e. temperature, pressure, wind speed and wind direction) were also continuously recorded for results to be recalculated to reference conditions (1013 hPa and 294°K) and expressed as a mass concentration in nanograms per cubic meter of air under reference conditions (Richard J: C. Brown et al., 2010).

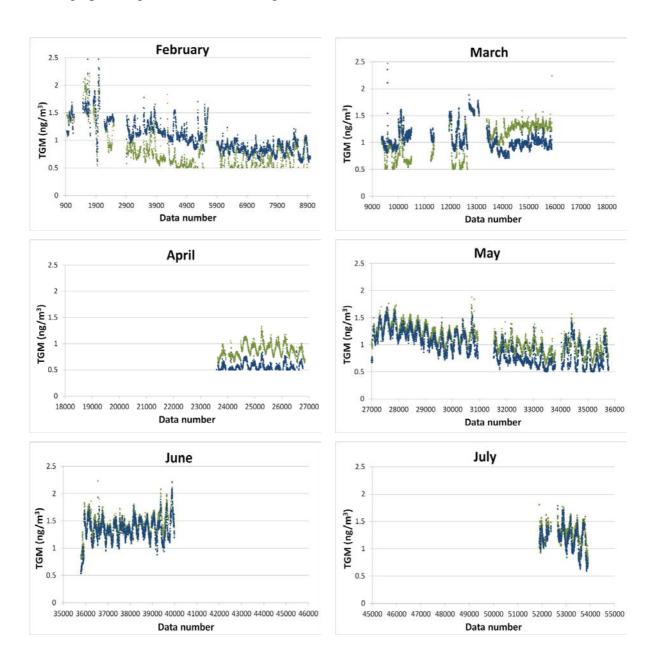
As quality control and quality assurance (QC/QA) the Tekran operation including the calibration test, the maintenance procedure and data validation were carried on according the Standard Operational Procedures established by GMOS. The documents were translated to Spanish (see Annex 1). Quarterly operational programs and monthly technical reports were doing as part of the QC/QA process (http://sdi.iia.cnr.it/gdqm/).

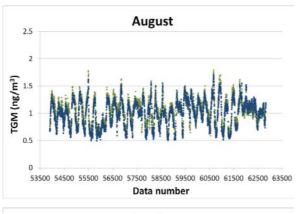
The descriptive statistics (maximum, minimum, median, mean, standard deviation), data distribution, and correlation tests (parametric and no parametric), between mercury vapor and meteorological parameters, were carried out with STATISTICA StatSoft program. The time series and wind roses were carried out using Caleida Graphic.

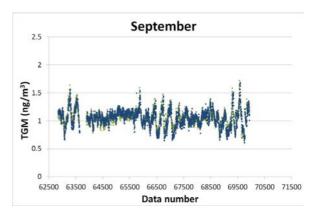
3. Results

3.1. Total gaseous mercury (TGM)

Total gaseous mercury (TGM) monitoring in air at the Celestún site (Gulf of Mexico coastal location) was initiated on January 28, 2012. Figure 5, shows the TGM data available at 10 m above ground from January to October 2012. TGM results displayed in the graphs ranged from 0.5 to 2.8 ng/m³.







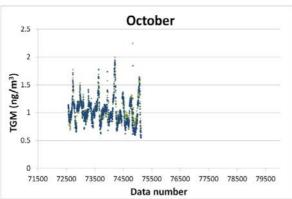


Figure 5. Time series of TGM concentration during 2012. Cartridge A (blue line) and Cartridge B (green line)

Due to technical and operational problems TGM data were obtained for 200 days during 2012. Annex 2, shows the annual time series of TGM concentration in continuous way. Initial operational problems were countered from February to April, where the measurement data displayed considerable differences between cartridge A and B. This problem was resolved by May.

For April, June, July and October frequent power break downs, delays in the delivery of gas and instrument's consumables (no Tekran dependence in Mexico) resulted in about 50% downtime of the instrumentation (Table 1).

Table 1. Operational problems

Period of time	Operational Problems						
from 24th March to 19th April	lack of argon gas in purge valve and delivering argon gas problems						
from 14th Jun to 25th July	Power failures, PC problems						
from 18th October to end of December	problems with the sampling pump						

Table 2 below, resumes the amount of available data, together with minimum, maximum and average TGM concentrations recorded per month and year. The annual TGM average obtained from 44537 data was 1.047 ± 0.271 ng/m³. The minimum value of 0.50 ng/m³ was registered from February to May; the lowest monthly average (0.752 ng/m³) was recorded in April. The maximum value (2.822 ng/m³) was observed in March, while the highest monthly average TGM value was obtained in June (1.388 ng/m³).

Table 2. Resume of all TGM values

	Data -	ng/m³								
Month	number (n)	Min	Max	Mean	Median	Standard				
	number (n)					Deviation				
February	5790	0.501	2.628	0.972	0.903	0.315				
March	4690	0.500	2.822	1.083	1.047	0.270				
April	2782	0.500	1.330	0.752	0.748	0.177				
May	7885	0.500	1.878	1.028	1.034	0.255				
June	4096	0.715	2.235	1.388	1.385	0.200				
July	1351	0.675	1.783	1.232	1.256	0.213				
August	8760	0.512	1.791	1.026	1.047	0.230				
September	6707	0.649	1.720	1.054	1.057	0.145				
October	2476	0.619	2.000	1.035	1.005	0.212				
Annual	44537	0.500	2.822	1.052	1.047	0.271				

The TGM average concentration reported in this study are similar to the levels reported in others locations consider as TGM background sites. Schmolke et al. 1999, reported TGM values in the range of 1.5 to 2.10 ng/m³ from four different monitoring sites located in the middle and northern of Europe. While in Canada was registered a mean value of 1.58 ng/m³ from 11 different rural locations considered remote from local sources (Temme et al. 2007). Reported TGM averages for 2004 for Mexico were around 1.46 ng/m³ at Puerto Angel (pacific coastal location in Oaxaca State) and about 1.32 ng/m³ at Huejutla (rural location in Hidalgo State) (De la Rosa et al., 2004).

Lindqvist et al. (1991) reported global background levels of 1.5 to 2.0 ng/m³ similar to those reported here, which suggest that Celestún location functions as a global TGM background site.

Although monthly TGM mean values displayed only discrete variations (table 2), the diurnal fluctuation of TGM were distinctive throughout the year for some months. The TGM diurnal patterns were generally characterized by highest TGM concentrations during nighttime compared to daytime. The peaks were registered from 3:00 to 6:00 local time, while lower TGM was observed from 14:00 to 17:00 local time. Figure 6 shows the inverse correlation between temperature and TGM content at Celestún measuring site. Kellerhals et al., 2003, indicated that diurnal fluctuations of TGM result from complex physical and chemical processes including: surface deposition, volatilization from surrounding surfaces, and transport. Several authors have been reported a TGM diurnal variation, found a highest TGM concentration during daytime (Zhu et al. 2012; Fu et al. 2008), while others have been registered highest values during nighttime (Lee et al., 1998; Schmolke et al., 1999). Schmolke et al., 1999, mentioned that the nighttime peaks of TGM concentrations could be attributed to mercury emissions from the surface accumulated in the nocturnal inversion layer.

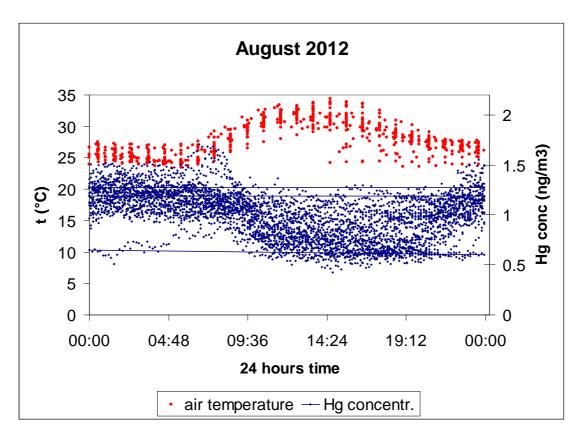


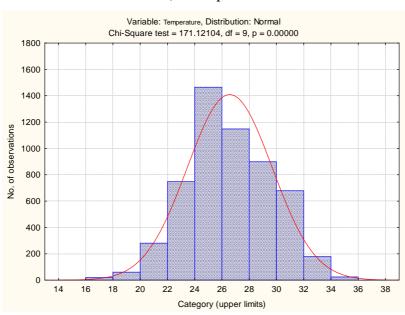
Figure 6. Temperature and TGM inverse correlation, (values for August 2012).

Celestún site is not a purely terrestrial site, because the influence of the Mexican Gulf has an impact on the air masses movements. Therefore temperature driven cycle of deposition and revolatilization from soil and vegetation is not the most remarkable phenomenon. At Celestún site, diurnal changes in wind direction, with nighttime winds from Southeast carrying terrestrial air masses and daytime winds, predominantly from the Northeast, carrying marine air masses will be present.

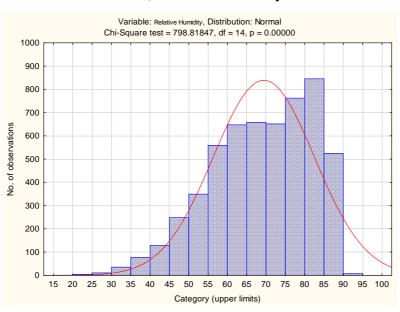
3.2. Meteorological Variables

Data distribution for the meteorological parameters of temperature, relativity humidity and pressure are shown in Figure 7.

a) Temperature



b) Relative Humidity



c) Barometric Pressure

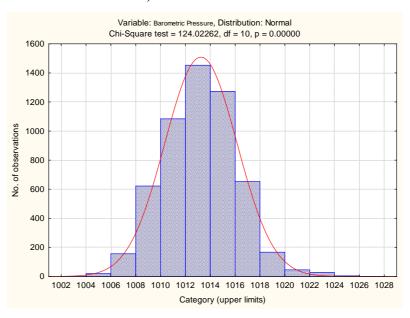


Figure 7. Data distribution; a) Temperature, b) Relative humidity and c) Barometric pressure.

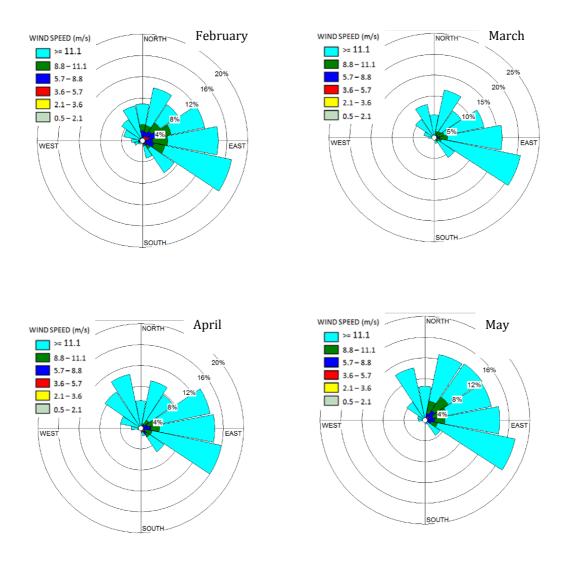
Table 3, shows monthly averages of the meteorological data registered during 2012. Temperature was of 26.57 °C, reaching temperatures at summer (May) around 27.4 °C. Average annual relatively humidity was 69%, and atmospheric pressure average was 1013 hPa. Rainfall was registered only at summer and autumn seasons.

Table 3. Descriptive statistic of the meteorological parameters

	Nº		Ter	nperature	(°C)		Barometric Pressure (hPa)				Relative Humidity (%)					
Month	Data	Min	Max	Median	Media	SD	Min	Max	Median	Media	SD	Min	Max	Median	Media	SD
February	684	17.10	33.70	24.05	24.26	2.96	1006.9	1023.1	1015.6	1015.4	2.68	30.00	91.00	75.00	72.49	12.90
March	647	17.60	34.10	25.30	25.64	3.46	1004.7	1024.7	1015.1	1014.6	3.89	24.00	89.00	64.00	62.21	13.77
April	713	17.40	35.30	25.90	26.11	3.16	1004.6	1019.1	1013.1	1012.7	3.04	27.00	91.00	64.00	63.92	13.37
May	738	21.80	35.80	27.00	27.35	2.66	1007.1	1015.8	1011.3	1011.2	1.67	30.00	91.00	70.00	68.92	11.27
June	419	22.70	35.20	26.80	27.36	3.16	1005.3	1015.5	1011.5	1011.3	2.10	36.00	91.00	74.00	72.30	13.66
July	226	22.50	33.30	26.60	27.21	2.82	1012.6	1019.1	1015.8	1015.7	1.31	45.00	90.00	79.00	74.34	12.13
August	743	23.30	34.40	27.50	27.90	2.69	1006.1	1016.8	1012.6	1012.4	1.96	37.00	89.00	74.00	71.51	12.48
September	704	23.40	33.50	27.15	27.37	2.60	1007.4	1017.1	1013.8	1013.6	1.70	40.00	89.00	76.00	73.41	10.89
October	630	16.80	32.20	26.10	26.40	2.61	1007.2	1018.7	1012.8	1013.1	2.71	40.00	88.00	71.00	69.72	11.95
Total	5504	16.80	35.80	26.30	26.57	3.11	1004.6	1024.7	1013.2	1013.2	2.91	24.00	91.00	71.00	69.42	13.09

Temperature, humidity and barometric pressure variation along the year at Célestun shows prevalence range (Q25 and Q75) of this parameters between 24.4 to 28.9°C, 60 to 80% and 1011 to 1015 hpa, respectively.

Monthly wind rose shows predominant wind from North-North-East (NNE) and South East (SE) from January to May. From June to September the dominant winds come mainly from the East and from October to November wind directions come from East and North East (NE) (Figure 8). Figure 8, also shows that the prevailing wind speed in Celestún site was equal or higher than 11 m/s. Soler-Bientz et al. 2010, reported at Celestún site high wind speeds (> 9 m/s), which attributed to the difference of temperature between the air masses over land and the sea surface.



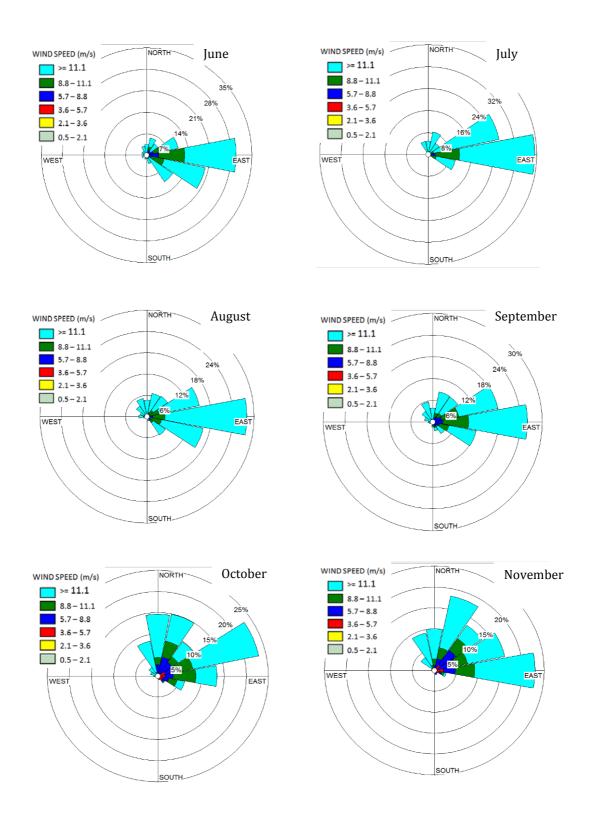


Figure 8. Monthly wind rose registered during 2012.

According, Figure 9 shows annual prevailing winds at Celestún coming from the land towards the West of Gulf of Mexico. Soler-Bientz et al. 2010, observed that at the

Yucatán Peninsula the wind come mainly from North-North-East (NNE) and East-South-East (ESE) where the easterly part of the Gulf of México and the westerly part of the Caribbean Sea are located. They also mentioned that the stability of the atmosphere over the sea influences the winds that arrive at the Yucatán Peninsula. It suggests that a possible contribution of TGM in the air of Celestún could be arriving from the Caribbean Sea and the east of the Yucatan Peninsula passing through cities as Mérida and Cancun.

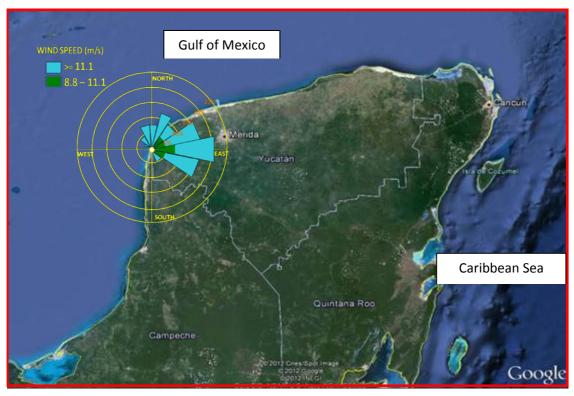


Figure 9. Average prevalent wind at the Celestún site for 2012

3.3. TGM and meteorological variables correlation

In order to evaluate the relation between TGM and meteorological variables an exploratory multivariate analysis was carried out. Cluster analysis (CA), with Ward's method and Pearson's correlation, a principal component analysis (PCA) and factor analysis were performed. Cluster analysis is a useful tool for classifying cases or variables into groups or clusters, where the degree of similarity is strong between members of the same cluster and weak between members of different clusters. The use of Ward's method in association with Pearson's correlation will ensure the highest

similarity between the members of a given group. Therefore, CA allows us to examine the grouping variables that act together in a given process.

At Figure 10, two groups are formed; the first one enclose TGM and relative humidity (RH), barometric pressure (BP) and rain precipitation (RP), while the second shows the formation of three subgroups, where solar radiation (SR), temperature (TEMP), wind and gust speed and direction (GS, WS, GD and WD) have no association with TGM, this grouping is further confirmed at Figure 11. Here TGM shows no direct relationship with air mass transportation from regional sources.

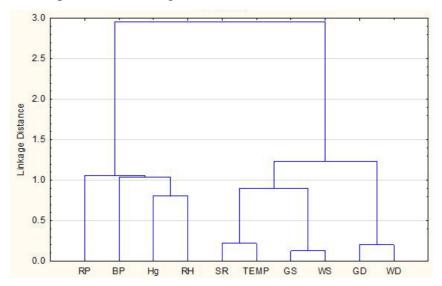


Figure 10. Cluster analysis for 10 variables. Ward's method with Pearson's correlation was used.

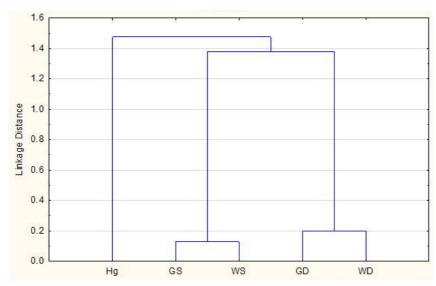


Figure 11. Cluster analysis for 5 variables. Ward's method with Pearson's correlation was used.

To further investigate on the relation between variables, a principal component analysis (PCA) with a varimax normalized rotation was also applied (Figure 12). This type of analysis maximizes the variances of the factor loadings across variables for each factor. Meaning these, that the generated graphic shows the best mathematical approximation for the real relation between variables which integrate a factor. The projection of the variables explain more than 55% of the observed variance, and it shows an horizontal axis representing the local atmospheric column processes (RH, PB, RP, temperature and SR) and the vertical axis with air mass transportation from regional sources (WD,WS, GD,WG).

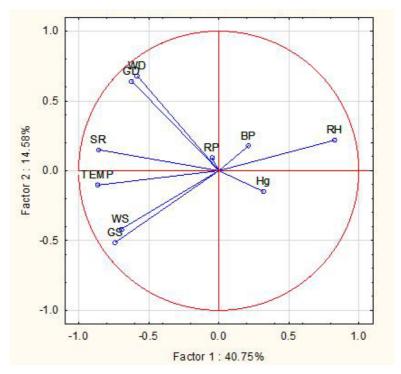


Figure 12. Projection of the variables on the factor-plane

To investigate the weight of each variable in the TGM variations, a factor analysis was performed. Three factors accounted for 64% of the total variance (Table 4). Factor 1 explains 40.7% of the total observed variance; it shows RH (loading of 0.827) in an inverse relation with almost all the variables, and a direct relation with TGM (loading of 0.310). This factor could be suggesting the diurnal variation influence on the TGM concentration. Factor 2, explains 14.5% of total observed variance, and shows that the significant variables are WD and GD, and they show an inverse non significant association with TGM (loading of -0.14), suggesting a non significant influence of those variables in TGM variation. Therefore, factor 2 could be suggesting that TGM variation

is independent of horizontal air mass transport from regional sources. Factor 3 explains 12.5% of the total observed variance and shows TGM (loading of 0.502), in an inverse relation with barometric pressure (-0.694). This indicates that vertical mixing of the atmospheric column could have a significant influence on TGM variations. A canonical analysis was performed to investigate the relation between the observed factors, and a significant relation (0.89) between factor 1 and 3 was found. This suggests that TGM variations could be mainly influenced by vertical fluxes of the atmospheric column related with diurnal variations.

Table 4. Factor analysis.

	Factor 1	Factor 2	Factor 3
WD	-0.58	0.67	0.27
GD	-0.62	0.64	0.25
WS	-0.69	-0.41	0.26
GS	-0.73	-0.51	0.18
TEMP	-0.86	-0.10	-0.22
RH	0.82	0.21	0.24
BP	0.21	0.17	-0.69
RP	-0.04	0.09	0.27
SR	-0.85	0.15	-0.29
Hg	0.31	-0.14	0.50
Expl.Var	4.07	1.45	1.25
Prp.Totl	0.40	0.14	0.12

4. Conclusions

The mercury levels registered in this study suggest that Celestún functions as a global TGM background site. The TGM range value registered in the study site was from 0.50 to 2.822, with an annual concentration average of 1.047 ± 0.271 ng/m³. Temperature, humidity and barometric pressure variation along the year at Celestún shows a range (Q25 and Q75) of these parameters between 24.4 to 28.9°C, 60 to 80% and 1011 to 1015 hpa, respectively. The predominant winds come from North-North-East (NNE)

and South East (SE) from January to May and in October to November wind directions come from East and North East (NE). The prevailing wind speed in Celestún site was up to 11 m/s

The correlation between TGM and meteorological variables suggested that TGM concentration could not have a direct relationship with air mass transportation from regional sources. The main correlation was found with the vertical mass distribution of the atmosphere by diurnal variation.

Due to the operational problem registered in Celestún site, that caused the loss of TGM data for a large period of time, we executed a relocation of the measurement station in a safer and more protected site. The new site chosen was the municipality of Sisal, around 40 km far from Celestún. The instruments were installed inside of the laboratories of the Facultad de Química, Unidad Sisal, of the Universidad Nacional Autónoma de México. The relocation to the new site was done in January 2013 and the instrument provides data since 25 January 2013.

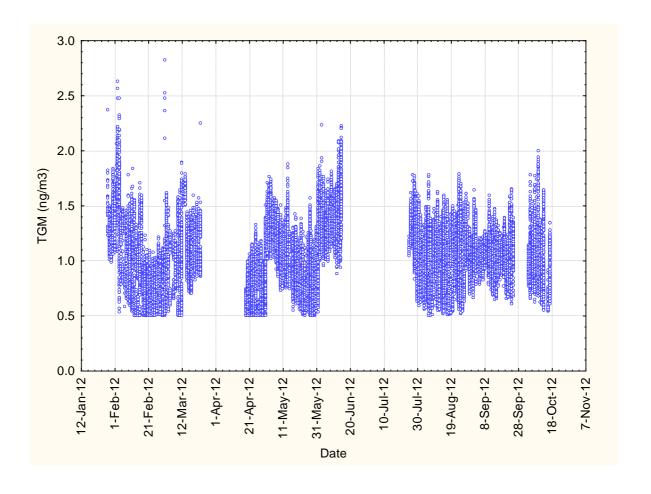
5. Annex 1. Procedures (in Spanish)

The following procedures are available for consulting or downloading at the JRC site:

ftp://ftp.ei.jrc.it/pub/support/GMOSProcedures

- 1. Procedimiento Estándar de Operación GMOS
- 2. Instructivos de operación y mantenimientos preventivos
- 3. Instructivo para subir los datos de las condiciones de trabajo del Tekran a la página web del GMOS.
- 4. Instructivo para graficar los datos en Excel

6. Annex 2. Annual Time Series



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Title: Wet deposition and atmospheric mercury monitoring in Celestún, Yucatán, México, as part of the Global Mercury Observation System (GMOS)

Author(s): Fabrizio Sena, Gunther Umlauf, Martha Ramirez Islas, Juan Antonio Velasco, Flor Arcega Cabrera, Ismael Oceguera Vargas

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Abstract

This report describes work conducted by the European Commission's Joint Research Centre in the contest of GMOS (Global Mercury Observation System). GMOS is an FP VII funded large-scale collaborative project aiming at the establishment of a Global Mercury Observation System including ocean-based, ground-based and atmospheric measurement activities under the umbrella of the GEO/GEOSS and the UNEP's Mercury program.

Within this 5 year project that started in 19 Nov 2010, JRC got the task to set-up a ground based station for measuring total atmospheric Mercury in Yucatán, Mexico. The system is fully automatic and transmits the acquired raw data via internet to the JRC for data analysis and evaluation prior to further reporting to the GMOS coordinator. Moreover, the time series obtained for Hg will be reported in the framework of Task Force on Hemispheric Transport of Air Pollution (LRTAP-Convention) co-chaired by the JRC (Climate change Unit).

In order to assist the JRC in the set up and routine maintenance of the Hg monitoring station, cooperation was initiated with the *National Institute of Ecology and Climate Change - Secretariat of Environment and Natural Resources (INECC-SEMARNAT)* in Mexico City and DUMAC foundation in Yucatán.

INECC agreed to assist the JRC in selecting an appropriate site partner, support maintaining the instrumentation, taking care of the wet deposition sampling/dispatch logistics, and supply meteorological data from the selected site.

Data regarding the total gaseous mercury concentration in air measured during the year 2012 at Celestún, Yucatán, México are here reported. The annual TGM average obtained from 44537 data was 1.047 ± 0.271 ng/m³. The minimum value (0.50 ng/m³) was registered from February to May, the lowest monthly average (0.752 ng/m³) was recorded in April. The maximum value (2.822 ng/m³) was observed in March, while the highest monthly average TGM value was obtained in June (1.388 ng/m³).

JRC Mission

As the Commission's in-house science service, the Joint Research Centre's mission is to provide EU policies with independent, evidence-based scientific and technical support throughout the whole policy cycle.

Working in close cooperation with policy Directorates-General, the JRC addresses key societal challenges while stimulating innovation through developing new methods, tools and standards, and sharing its know-how with the Member States, the scientific community and international partners.

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