ACCELERATOR BASED FACILITY DEDICATED TO AEROSOL POLLUTION RESEARCH IN MEXICO CITY

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An accelerator based ion beam facility dedicated to atmospheric aerosol pollution research has been developed at the Instituto Nacional de Investigaciones Nucleares (ININ), Mexico. Since atmospheric pollution in the Mexico City conurbation has become a prime concern for citizens, governmental authorities, decision makers and health and environmental researchers, new and precise information on atmospheric aerosol pollutants is required to understand better and eventually control and reduce it. Accelerator based ion beam analysis techniques such as PIXE have shown to be very powerful to determine aerosol elemental content and to provide basic information to identify aerosol pollution sources. A PIXE beam line, with the majority of its elements being locally built, was designed to match a new 2MV Tandetron accelerator facility and it has been dedicated to aerosol pollution analyses. The facility is nowadays fully operational and currently provides information to governmental authorities, decision makers and to health and environmental care institutions in the country. This information is complementary to the results obtained by other research institutions dedicated to monitor air pollution in Mexico City. The facility is also intended to provide analyses of aerosol samples from other conurbation areas in the country in the near future.

I. INTRODUCTION

Air pollution studies in heavily populated cities are nowadays more relevant due to the increasing health risks to which human beings are exposed. Local authorities, citizens, and research groups working on health effects and health risks assessment of air pollution continuously demand this information. Thus, these studies are expected to provide the most comprehensive information possible. As it is well known, ion beam based analyses of airborne particulate matter have shown great capability to provide the required reliable information. These facts have encouraged scientists working with accelerator machines to commit themselves to this promising research field adapting more facilities to this purpose, especially those of low energy output. PIXE studies started at ININ (Mexico) during the early nineties using a 6 MV tandem Van de Graaff accelerator formerly dedicated to nuclear physics research only. A PIXE set-up intended to perform studies of airborne particulate matter was installed[1,2] and its first results delivered to the community. Nevertheless, since this installation is highly demanded by several research groups, the machine time available to the PIXE studies was scarce and therefore a new 2 MV accelerator was installed by the year 1999. It reached operational status by the middle of the year 2000, and helped indeed to alleviate that inconvenience. A view of the new facility is shown in Fig. 1.

A new PIXE line was also implemented and put into operation soon after. At present, the facility is dedicated to perform analyses of airborne particulate matter, using PIXE as the basic analytical technique. However, in order to take full advantage of this machine, new lines are under consideration. An outline of the ongoing research program is presented, and some comments on the research prospect to be developed at ININ in the near future are also given.

II. THE ININ 2 MV TANDEM VAN DE GRAAFF ACCELERATOR FACILITY

II.A. The Necessity of a New Ion Beam Facility

In 1998, ININ acquired a 2 MV Tandem Van de Graaff accelerator model 4120 MC from High Voltage Engineering Europa B.V. The Mexican Treasure Secretariat (Secretaría de Hacienda) authorized the funds for the new machine, urged the necessity to enforce continuous environmental research in large urban areas of Mexico, especially in the Mexico City's Metropolitan Area (AMCM), recognized as one of the most polluted throughout the world. This decision was made firmly based on the great capability shown by the IBA techniques to determine environmental pollutants in a fast and reliable way. The ININ-PIXE group has provided results[3-10] on atmospheric pollution in the AMCM and other locations of the Mexican territory for several years. The results obtained constitute a reliable source of information on atmospheric pollution not only for the production scientific publications, but also as a tool of social policy decision making.[7]

Fig. 1. The 2 MV ion beam accelerator facility at ININ, Mexico.

The capability for PIXE analysis at ININ was established[1,2] at the beginning of the nineties, when the first PIXE installation was completed and the system was calibrated and validated. Since then, several publications[2-14] have been issued presenting results aimed at alleviating the acute atmospheric pollution problem in the AMCM. The increasing demand for information about atmospheric pollution, fundamental to better understand and attempt to solve this problem, has created a real need of fresh results in the field. The new ININ PIXE beam line followed the installation of a 2 MV Tandetron accelerator, and it provides a substantial part of the information required in the country. Most of the components of this set-up, with the exception of a few very specialized ones, were locally built and the whole assembly has shown an excellent performance. Details of the experimental array and of the system calibration are given elsewhere. [15] The validation of results was carried out comparing those from a sample set previously analyzed at the Crocker Nuclear Laboratory of the University of California, Davis, versus the analyses of the same set using the ININ facility. Following the calibration and validation process, current aerosol samples collected in the AMCM have been analyzed as part of an ongoing collaborative research project ongoing between ININ and the Air Environmental Management Secretariat of Mexico City's local government (Dirección General de Gestión Ambiental del Aire del Gobierno del Distrito Federal, DGGAA-GDF).

II.B. Installation Process

The machine, received at ININ in April 1999, was installed in a remodeled building fulfilling most of the requirements at the arrival time. A few aspects of the installation process are pointed out in the following lines, as a reference for research groups involved in such a delicate task. In the first place, fulfilling all installation requirements is fundamental indeed. This action prevents possible problems that may be encountered during the installation process. Secondly, it is highly recommended to support the facility with an uninterruptible power supply (UPS) system. Although this is not an indispensable requirement of the accelerator manufacturer, it can keep the machine running under unexpected power failures lasting up to few minutes, saving the settings of the electronics equipment as well as protecting the facility from surge transients and overload charge. This investment pays in the short term and it is highly recommended over an alternate emergency power plant, useful only during longer power failures, which has several drawbacks in protecting the equipment. Finally, the installation of a fast closing valve to protect the accelerator from abrupt vacuum breakings is highly recommended. This valve ought to be inserted into the beam transport pipe, just between the accelerator tank and the switching magnet, in order to protect all lines at a time. All couplings and fittings required to install it were locally built at ININ's workshops, showing excellent performance. Fig. 2 shows the fast valve installed into the beam line. The installation was accomplished by June 2000. Once the facility reached operational status, a PIXE line was immediately installed.

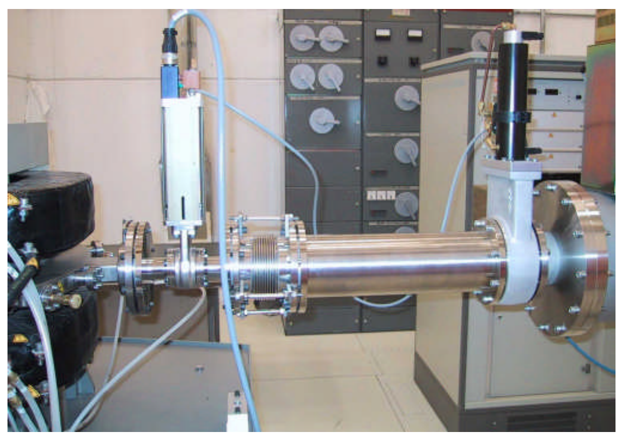

Fig. 2. Fast valve inserted into the beam line. All couplings were locally built.

II.C. The New PIXE Beam Line

In accordance with the dimensions of the accelerator room, which was adapted to install the new machine, it was decided to install the PIXE beam line in the +10º position. This allowed enough space to place the irradiation chamber at an appropriate distance from the switching magnet, the latter device having four exit ports. The target position was located at 5.40 m from the 10º exit port.

The locations of some of the most important elements placed between the exit port and the target chamber are: i) a pneumatic valve is placed just after the exit port. ii) a couple of magnetic dipoles are installed in order to provide a final steering of the ion beam and to optimize the beam transmission. These steerers were used mainly during the test runs and few of the normal bombardments, because it was possible to focus the beam directly with the high energy focusing system of the accelerator. iii) the chamber containing a diffuser foil is located 4.4 m downstream from the exit port, and one meter before the target. This chamber is outfitted with water-cooled current scrapers both at its entrance and exit, a quartz, a Faraday cup, and the 6 µm Al diffuser foil. The latter placed 4.4 m downstream from the exit port. The scraper at the exit is, in fact, a double one, with the first attached at 30 mm, and the second at 88 mm from the diffuser foil. This close position is aimed to prevent the beam to spread too much and also to define more precisely the size of the beam transported to the target. The double scraper is located inside a coupling element that connects the diffuser foil chamber and the irradiation chamber. At the end of this coupling element there is a third scraper, which defines the final size of the beam transported to the target. iv) the irradiation chamber is placed with the target located 5.4 m downstream from the exit port. A view of the PIXE beam line is shown in Fig. 3.

Fig. 3. PIXE line coupled to the ion beam accelerator facility at ININ, Mexico.

Several improvements were made in the new PIXE set-up in with respect to the previous one. A few of these are: The set of aluminum beam-defining apertures, originally installed in the irradiation chamber by the manufacturer, was replaced by a new set made of tantalum, in order to reduce undesired radiation to a minimum. In this action, the diameter of the apertures was increased to 5.0 mm. This could be done because the ion beam is more uniform and more stable in the new PIXE installation than in the former one. This modification permitted a reduction of the average sample irradiation time, down to around one third of the original time, without diminishing the quality of the spectra. A new detector mount, fitted with a tri-directional fine adjustment system as the one formerly used[1] in the first PIXE set-up, was designed to facilitate an accurate repositioning of the detector, contributing to a better experimental reproducibility. The whole assembly, exhibited in Fig. 4, has shown excellent performance.

Fig. 4. PIXE arrangement installed at ININ, Mexico.

During the first stage of analyses carried out after the PIXE line was built, the characteristic X-rays were collected using a Si-Li detector with a resolution of 210 eV at 5.898 keV, and a 6 mm diameter active area. A suitable lead collimator, meant to reduce detector edge effects limited the detection area. Samples to be analyzed are placed at a 45º angle with respect to the ion beam direction. The port for the X-ray detector is located at a 90º angle with respect to the same direction. Typical bombardment particles are 2.0 to 2.5 MeV energy protons intended to obtain the best overall sensitivity. A funny filter with a suitable aperture is usually installed to balance the detection of low and high energy X-rays. Different funny filters are available in the laboratory in order to choose the appropriate one to achieve the required balance. Recently, a new HPGe detector with much better resolution, 145 eV at 5.898 keV and an active area of one square cm, has been installed to replace the Si-Li detector.

The system is currently used to perform PIXE analyses of airborne particulate matter samples from the AMCM. It is expected that the results obtained be widely used, in the short term, among research groups and local authorities responsible for atmospheric pollution issues. It is also common analyzing samples from other urban areas of the Mexican Republic, and even from some other Latin-American countries.

II.D. System Calibration and Validation of Results

The calibration of the PIXE system was performed using spectroscopically pure thin samples of known areal density (30-70 μg/cm^2) obtained from Micromatter®. These thin samples have a quoted accuracy of ±5% and are deposited on polycarbonate substrates. Twenty-seven suitable pure samples were selected to cover the range of elements of interest, 22 for K X-rays and five for L X-rays. The X-ray yields per μC were determined for each calibrator for both K and L X-rays, and two calibration curves, shown in Fig. 5, were obtained plotting X-ray yields per μC per μg/cm^2 as a function of atomic number Z. In this calibration process, the number of pure samples was increased in comparison with the calibration performed in the old facility, by including five elements of the L X-ray series.

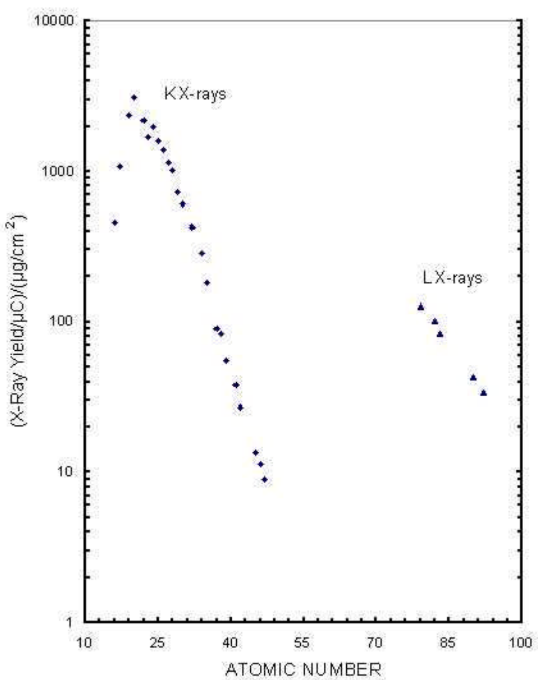

Fig. 5. K and L X-ray calibration curves of the new PIXE line.

The precision of the process was determined[2] by making repeated measurements under constant conditions on a given thin film. The standard deviations obtained for each group of measurements for a given element, which are estimations of the precision, are in most cases 1-5% of the mean, and are in good agreement with each other. A set of samples previously analyzed at the Crocker Nuclear Laboratory of the University of California, Davis, was analyzed anew to validate the results. The analyses of several well-preserved aerosol samples, which had been analyzed some time ago in the old ININ PIXE system, were also performed using the new one, and the comparison of the old and new spectra attested the results obtained.

The uniformity of the ion beam was measured using a special aluminum target with a tiny chip of copper in the center of the frame. The ion beam profile was obtained scanning the beam cross section, of about 5 mm diameter, by irradiating this target at many different positions separated by small steps. It was found that the beam profile was uniform within 4 % over its cross sectional diameter, as can be seen in Fig. 6. The uniformity of the beam appeared considerably improved with respect to that obtained in the old facility.

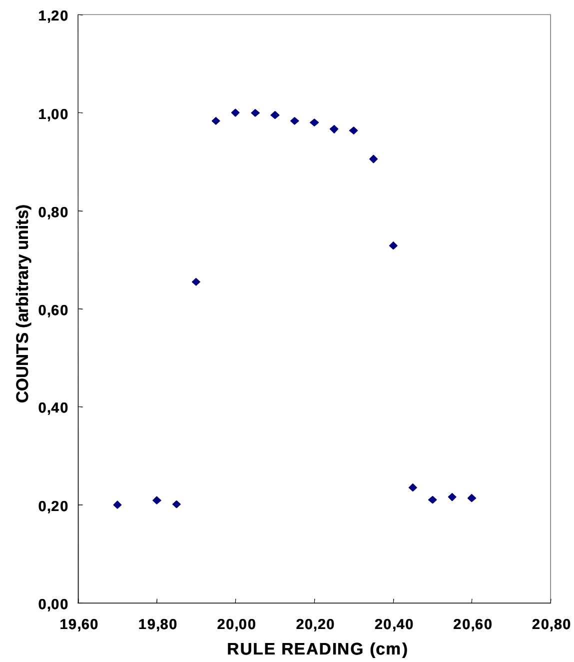

Fig. 6. Beam profile uniformity of the irradiation area.

III. RESEARCH AND APPLICATIONS

III.A. The Research Ongoing at ININ

As mentioned before, atmospheric pollution studies have been performed at ININ for over one decade and, since the year 2000, using a 2 MV Tandetron accelerator. At the moment, two main projects comprise the ongoing research activities, both related to atmospheric pollution. The first one is concerned with the determination of elemental content in airborne particulate matter (APM) samples collected in the AMCM. In this project, conducted in collaboration with the DGGAA-GDF, the main $PM_{2.5}$ APM pollution sources have been identified[14] and their corresponding source apportionments were also determined. The results obtained were found in good agreement with the emissions inventory[16] carried out by the local government. It is worth mentioning that pollution source identification is much easier thanks to the information that can be obtained from the PIXE analysis of the samples, from which a relevant database can be obtained. This database must be large enough to be statistically significant under any of the statistical methods commonly used to identify and determine the source apportionment contributions of the main pollution sources. The information provided by PIXE is fundamental to perform such statistical analysis. Results of the mean source contributions to fine elemental concentrations determined for the PEDREGAL site[13] of the AMCM are shown in TABLE 1. The second project, funded by the council of science and technology of the Mexico State (Consejo Mexiquense de Ciencia y Tecnología del Estado de México, COMECyT), is focused on $PM_{2.5}$ APM characterization in downtown Toluca, the state capital, a multi-faceted city located 65 kilometers west of Mexico City which nowadays counts nearly one million inhabitants in its conurbation. Many industries are located in the surroundings and many thousands of internal combustion vehicles circulate in the area everyday. In this project, fine particle aerosol samples are collected, every third day since March 2006, in a sampling site located in downtown Toluca, using an EPA $PM_{2.5}$ reference method sampler. The samples collected are currently PIXE analyzed in order to determine their elemental content. The database derived from these analyses will be used to identify the local $PM_{2.5}$ APM sources and the corresponding source apportionments. This is the first estimation of fine particle air pollution performed in this area ever.

The results obtained in both projects form part of the information used by local authorities responsible of making decisions on atmospheric pollution issues. Moreover, some research groups conducting studies on health effects and health risk assessment of atmospheric pollution have already requested results of the ongoing studies. It should also be mentioned that some other studies performed in the recent years using this facility, have also provided valuable information for several state and municipal governments. Thus, updated regulations on atmospheric pollution, local and national, have taken into account the overall information generated by the ININ PIXE group. The PIXE laboratory also provides APM PIXE analyses to other countries in Central America and the Caribbean, as well as training scientists and technicians of the region.

TABLE 1. Mean source contributions to fine elemental concentrations. Site: PEDREGAL.

Variable	Soil	Small industry	Sulphates plus FPM	Incinerators and other burnings	Sum of estimated mean contributions	Observed mean concentrations	Observed estimations r^2
FPM	2.91	1.68	6.80	5.51	16.90	17.53	0.96
S	0.00	164.30	873.29	244.37	1281.95	1372.54	0.93
Cl	16.25	11.81	0.00	52.69	80.75	84.99	0.95
K	22.33	13.19	25.07	22.80	83.38	81.34	1.03
Ca	59.61	12.15	19.00	21.40	112.16	116.47	0.96
Ti	7.96	0.91	2.17	3.11	14.15	14.35	0.99
Mn	3.78	1.44	0.00	1.53	6.75	7.33	0.92
Fe	92.86	16.05	12.04	24.62	145.57	148.28	0.98
Ni	0.47	0.35	1.70	-0.47	2.04	2.17	0.94
Cu	1.17	4.00	2.13	1.42	8.72	8.94	0.98
Zn	7.61	38.54	0.00	18.12	64.28	67.97	0.95
Br	0.85	0.00	1.02	4.94	6.80	7.21	0.94
Pb	1.61	12.27	4.92	0.00	18.81	26.49	0.71

Since the new facility was put in operation, several research collaborations have been undertaken both with local and international counterparts. It is worth mentioning the Study of APM collected in big Latin American cities, a regional project conducted with researchers from Argentina, Brazil and Chile. Likewise, the national project 'Study of Fine Particle Contamination in Mexico City' has been conducted in collaboration with DGGAA-GDF, both funded by the International Atomic Energy Agency (IAEA). Other projects continue in progress.

III.B. The Research and Development Prospect

Research on atomic and nuclear fields at ININ has been conducted continuously for over 30 years. During this period of time, a good scientific and technical capability has also been developed. Human resources have been prepared as well, which led to a significant participation in the country's development and has also contributed to the welfare of the population. Among these collaborations, those aimed to the solution of problems related with environmental issues have been indeed significant. The PIXE research group has had a direct participation in establishing the limits of maximum permissible levels of contaminating particles in the atmosphere as well as some polluting elements such as sulfur, manganese and lead. Most of this research has been performed using atomic and nuclear techniques based on the use of ion beam accelerators. The new ion beam facility developed at ININ will contribute significantly to alleviate the lack of resources in order to continue the research activities already in progress. This will also permit the use of complementary techniques, which, although well known by the scientific community, have not been implemented at ININ yet. The research and development prospect for the new accelerator includes four lines of research, which should be completed in order to make the best of the investment done on the new accelerator. The proposed lines are: 1. The PIXE line to analyze atmospheric aerosol APM samples, already in an operational status. 2. An external beam to enable the analyses of biological specimens, archaeology and art pieces, and soil samples. The benefits of having this line will pay in the short term. 3. A line to do research into materials sciences including materials modification. 4. This line may well be dedicated to educational and training purposes. Multiple experiments could be conducted in it, allowing ININ to train its own personnel as well as students from diverse universities.

Recently several Latin-American countries have proposed to the IAEA to designate the ININ PIXE laboratory as a Regional Training and Research Center for Latin America and the Caribbean, having it as the key laboratory to perform atmospheric pollution research using IBA techniques. This proposal is currently under assessment. It is expected the agency to provide the required funds to keep this project in operation.

IV. CONCLUSIONS

The new ion beam facility developed at ININ has been successfully installed and it is in operational status. Four lines of research have been proposed to exploit in full the potential of the new machine: 1. The PIXE line for atmospheric research studies, already in use. 2. An external beam line for biological and soil samples, archaeometry and art pieces. 3. A line for materials research. 4. A line for educational and training purposes. The benefits that these settings can bring to the society are promising. At the present stage, the facility has enabled to better understand the atmospheric pollution problem in large urban areas of Mexico and other countries in Latin America.

The PIXE facility built at ININ is nowadays fully dedicated to perform important environmental research studies, particularly APM analyses. It has been evaluated both in terms of precision and reproducibility with very satisfactory results. The elemental content in APM determined with this system has been validated by comparing results with accurate measurements previously performed on two other different systems. This has demonstrated that the protocol adopted at ININ for the measurement of elemental content in APM using PIXE is satisfactory. Thus, it is possible to conduct absolute mass elemental concentrations of atmospheric aerosol samples with confidence, with a considerable reduction in irradiation time per sample. The facility has shown very good overall performance, providing reliable information, sometimes difficult to be obtained by means of other analytical techniques, which can be used by local authorities, researchers, health institutions and decision makers.

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REFERENCES

1. J. FLORES M., F. ALDAPE, R.V. DÍAZ and D. CRUMPTON, *Nucl. Instr. and Meth.* **B75** 116 (1993).
2. F. ALDAPE, J. FLORES M., R.V. DÍAZ and D. CRUMPTON, *Nucl. Instr. and Meth.* **B75** 304 (1993).
3. F. ALDAPE, J. FLORES M., R.V. DÍAZ and D. CRUMPTON, *Nucl. Instr. and Meth.* **B109/110** 459 (1996).
4. F. ALDAPE, J. FLORES M., R.GARCÍA G. and J.W. NELSON, *Nucl. Instr. and Meth.* **B109/110** 502 (1996).
5. F. ALDAPE, and J. FLORES M., *Int. J. of PIXE*, **6** 205 (1996).
6. R. URIBE-HERNÁNDEZ, A.J. PÉREZ-ZAPATA, J. FLORES M., F. ALDAPE, and B. HERNÁNDEZ-MÉNDEZ, *Int. J. of PIXE*, **6** 255 (1996).
7. F. ALDAPE, B. HERNÁNDEZ-MÉNDEZ and J. FLORES M., *Nucl. Instr. and Meth.* **B150** 363 (1999).
8. F. ALDAPE, J. FLORES M., R.V. DÍAZ, B. HERNÁNDEZ-MÉNDEZ, J.M. MONTOYA Z., E.E. BLANCO, A.F. FUENTES and L.M. TORRES-MARTÍNEZ. *Nucl. Instr. and Meth.* **B150** 439 (1999).
9. J. FLORES M., F. ALDAPE, R.V. DÍAZ, B. HERNÁNDEZ-MÉNDEZ and R. GARCÍA G., "PIXE analysis of airborne particulate matter from

Xalostoc, Mexico: winter to summer comparison," *Nucl. Instr. and Meth.* **B150** 445 (1999).
10. J. FLORES M., and F. ALDAPE, "PIXE study of airborne particulate matter in northern Mexico City," *Int. J. of PIXE*. **11** 61 (2001).
11. R.V. DÍAZ, F. ALDAPE, and J. FLORES M., "Identification of airborne particulate sources of samples collected in Ticomán, Mexico, using PIXE and multivariate analysis," *Nucl. Instr. and Meth.* **B 189** 249 (2002).
12. F. ALDAPE and J. FLORES M., "Analysis of airborne particulate matter collected after an eruption episode of the Popocatépetl Volcano," *Int. J. of PIXE* **13** 133 (2003).
13. F. ALDAPE and J. FLORES M., "Source apportionment of fine airborne particulate matter collected in the Mexico City Metropolitan Area," *Int. J. of PIXE* **14** 147 (2004).
14. F. ALDAPE, J. FLORES M., J. FLORES A., A. RETAMA-HERNÁNDEZ and O. RIVERA-HERNÁNDEZ, "Elemental composition and source identification of $PM_{2.5}$ particles collected in downtown Mexico City," *Int. J. of PIXE* **15** 263 (2005).
15. F. ALDAPE, "Identificación de principales fuentes contaminantes de partículas finas en la Ciudad de México: Uso de rayos-X inducidos por bombardeo de protones y métodos estadísticos," Ph. D. Thesis, Universidad Autónoma Metropolitana, U. Iztapalapa, (2006).
16. In: Inventario de Emisiones a la Atmósfera, Zona Metropolitana del Valle de México, Secretaría del Medio Ambiente del Gobierno del Distrito Federal, Published by SMA-GDF, Mexico City, Mexico (2000) 41.