

**INFLUENCE OF ANTHROPOGENIC SOURCES ON TOTAL GASEOUS MERCURY
VARIABILITY IN GRENOBLE SUBURBAN AIR (FRANCE)**

**Aurélien Dommergue¹, Christophe P. Ferrari^{1,2,*}, Frédéric A.M. Planchon¹ and Claude F.
Boutron^{1,3}**

¹ Laboratoire de Glaciologie et Géophysique de l' Environnement du CNRS, 54 rue Molière, BP 96,
38402 Saint Martin d'Hères, France.

² Institut des Sciences et Techniques de Grenoble, Université Joseph Fourier, 28 Avenue Benoît
Frachon, BP 53, 38041 Grenoble, France.

³ Unités de Formation et de Recherche de Mécanique et de Physique, Université Joseph Fourier
(Institut Universitaire de France), BP 68, 38041 Grenoble, France.

Corresponding author. *Tel.*: +33 4 76 82 42 00; *fax*: +33 4 76 82 42 01

E-mail address: ferrari@glaciog.ujf-grenoble.fr

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Abstract

Total Gaseous Mercury (TGM) has been monitored at Champ sur Drac, a suburban site of Grenoble in southern east France. TGM measurements have been made over 4 periods of about 10 days throughout 1999-2000 using cold vapour atomic fluorescence absorption technique. The first monitoring campaign was initiated on November 4, 1999, followed by three other campaigns respectively on January 12, 2000, April 10, 2000 and July 17, 2000. Concurrent monitoring of O₃, NO, NO₂, SO₂ and of meteorological parameters have also been performed. The mean TGM concentration was 3.4 ng m⁻³ with maximum hourly mean concentration of 37.1 ng m⁻³. Although mean TGM concentration was not greatly different from those previously measured in the

troposphere, the greater TGM variability as well as the occurrence of high TGM concentration linked to particular wind conditions suggested the strong influence of anthropogenic sources. The chlor-alkali plant located nearby, the others chemical industries using fuel combustion and the municipal waste incinerator were thought to contribute to mercury pollution events.

Keywords: Total Gaseous Mercury; Atmosphere; Variability; France; Anthropogenic sources

1. Introduction

Since the pollution prevention concept emerged many years ago (Pacyna, 1986), there has been a growing concern about air quality in urban areas. To improve urban air quality, environmental policy makers expressed widespread interest in controlling and regulating major gaseous pollutants such as O₃, CO, NO, NO₂, SO₂ and VOC (Volatile Organic Compounds). However gaseous mercury supervision is only from time to time included in urban pollution monitoring whereas the knowledge of its distribution and transport pathways are of critical importance. Enlarging air pollution monitoring program to atmospheric gaseous mercury survey can only ensure this.

Gaseous elemental mercury (Hg⁰) is the dominant form (~ 98% *e.g.* Poissant, 2000) of mercury in the atmosphere. Elemental mercury is very insoluble, unreactive (Schroeder and Munthe, 1998, Tokos et al., 1998) and thus has a residence time in the atmosphere of about 0.5-2 years (Lindqvist and Rodhe, 1985; Slemr *et al.*, 1985; Munthe and McElroy, 1992). Therefore Hg⁰ is well mixed atmospherically and Total Gaseous Mercury (TGM) concentration is in the range 1-5 ng m⁻³ in the troposphere (Lee *et al.*, 1998; Ebinghaus and Slemr, 2000; Urba *et al.*, 2000).

Mercury is emitted into the atmosphere from a variety of natural and anthropogenic sources. The natural sources include volcanoes, crustal degassing, forests, lakes and oceans (Lindqvist and Rodhe, 1985, Mason et al., 1994), while fossil-fuel combustions, incineration, metallurgical processes and chlor-alkali plants constitute the largest part of anthropogenic Hg emissions (Pirrone

et al., 1996). In French urban areas, waste incineration is the major source of Hg (CITEPA, 2000). The other contributions are coal combustion followed by chlor-alkali plants, wood combustion and metallurgic processes. Once emitted, mercury is involved in many physical and chemical transformations (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999) and can be deposited by wet and dry processes to environmental surfaces. These deposition processes are more efficient if mercury is in the +II oxidation state (Lindberg and Stratton, 1998).

Since information on background concentrations and anthropogenic sources are still lacking for many places in Europe, we present here the first automated long-term, high time resolution measurements of TGM concentration in France. To our knowledge, this report constitutes the first presentation of extended atmospheric mercury data from a site in France. We attempted to compare these data with other pollutants (O_3 , NO, NO_2 , SO_2) concentrations and meteorological parameters (wind speed, wind direction, atmospheric pressure, temperature, solar irradiation and precipitation intensity). An assessment of potential anthropogenic mercury sources and their influence on our measurement site are also provided.

2. Material and methods

2.1 Site characteristics

TGM concentrations were determined in the vicinity of Grenoble in the south east of France (Fig. 1). The measurement station is located at Champ sur Drac (45.080° N, 5.730° E, 267 meters a.s.l.) which is a suburban site of Grenoble, characterized by an industrial zone in the north and particularly a chlor-alkali plant (Fig. 2) using mercury cell process. Grenoble (~400,000 inhabitants) is ~10 km north from the site and has one of the largest municipal waste incinerator in the area.

2.2 Sampling and analysis

TGM measurements were performed during four ~10-days campaigns. The first campaign was initiated on November 4, 1999. Others measurements followed respectively on January 12, 2000, April 10, 2000 and July 17, 2000.

Air samples were automatically collected outside with a Teflon line at 3 meters above the ground. We initiated routine measurements with an automated Gardis 1A+ analyser (Ekoservis, Lithuania) using the double amalgamation technique followed by Cold Vapour Atomic Absorption Spectrometry (C.V.A.A.S.). During each campaign TGM concentrations were measured every 15 minutes, 24 hours a day. The analyser, housed in air-conditioned shelter, was programmed to sample air at a flow rate of 1 l min⁻¹. Particulate matter was removed by 25-mm diameter Teflon filter (0.5 µm). The performances of the Gardis analyser for TGM monitoring have already been verified successfully through atmospheric measurements (Urba *et al.*, 1995; Tan *et al.*, 2000; Urba *et al.*, 2000) and international intercomparison exercises (Ebinghaus *et al.*, 1999; Munthe *et al.*, 2001). Full details concerning accuracy and precision of our instrument can be found elsewhere (Ferrari *et al.*, 2000) and calibration procedures and curves are given in Ferrari *et al.* (2001). A detection limit of about 0.1 ng m⁻³ was achieved.

Moreover concurrent monitoring, recorded every 15 minutes, of meteorological parameters (wind speed, wind direction, atmospheric pressure, temperature, solar irradiation and precipitation intensity) and of O₃, NO, NO₂, SO₂ facilitates the interpretation of Hg data.

3. Results and discussion.

3.1 Annual, seasonal and diurnal variability of TGM

A statistical summary of TGM data subdivided into their respective seasons is presented in Table 1. The mean seasonal TGM concentration varies throughout the year from 1.9 ng m⁻³ to 4.8 ng m⁻³. The mean TGM concentration (plus 1 SD) calculated over the year is 3.4 ± 3.6 ng m⁻³ (N = 3596). Maximum TGM concentration was recorded in July (45.9 ng m⁻³) with an hourly mean concentration of 37.1 ng m⁻³ for this event.

Ozone mean concentrations are ranging from ~7 ppbv for winter data to ~30 ppbv for summer data with peaks up to ~70 ppbv. In southern Quebec, Poissant (1997) observed some evidence of chemical oxidation of TGM by ozone under high water vapour mixing ratio and ozone concentration higher than 30 ppbv. In Champ sur Drac, no significant correlation was noticed between TGM and ozone values. One cannot conclude that there is no oxidation of Hg⁰ by ozone. It rather confirms that this reaction is slow and that reactions in urban atmosphere are various and complex.

The average TGM concentration for each season as well as for the entire set of data are comparable to those typically measured in the troposphere (*i.e.* in the range 1-5 ng m⁻³, *e.g.* Lee *et al.*, 1998; Ebinghaus and Slemr, 2000; Urba *et al.*, 2000). Nevertheless, as regarding the temporal variability of 15 min TGM data (Fig. 3), one can notice that under particular wind conditions a strong influence of anthropogenic sources of Champ sur Drac and Grenoble is observed. Although low TGM concentrations (around 2 ng m⁻³) are the most recurrent, there are many peaks above 10 ng m⁻³ in each campaign (approximately 10 % of fall and summer data).

There are limited published data in France with which to compare our measurements. It is thus difficult to address a precise comparison regarding the Hg levels in French cities. Nonetheless, Hg⁰ has already been determined in a few air samples in Bordeaux (Pécheyrat *et al.*, 2000) using a different technique. They found mean Hg⁰ concentrations of 2.7 ng m⁻³ and 4.0 ng m⁻³ in open urban area and suburban sites respectively. However, the number of samples collected was limited and inadequate to discuss on TGM variability.

On the other hand, several atmospheric TGM monitoring campaigns have been already achieved in the rest of the world. Measurements made in clean atmosphere have shown relatively similar pattern with concentration of the order of 1.5 ng m^{-3} and with a variability of low amplitude. Indeed, in the atmosphere over the southern Baltic Sea coast, *Urba et al.* (2000) reported mean TGM concentration of 1.37 ng m^{-3} and 1.94 ng m^{-3} (in the range $0.68\text{-}2.20 \text{ ng m}^{-3}$ and $0.98\text{-}3.81 \text{ ng m}^{-3}$ respectively). *Schroeder et al.* (1995), during the first winter cruise of the "Polarstern" from Germany to the Nordic Seas recorded a mean concentration of 1.47 ng m^{-3} ($0.67\text{-}2.82 \text{ ng m}^{-3}$). Measurements performed at the summit of Wank mountain in Germany (*Slemr and Scheel, 1998*) showed data ranging from 1.80 ng m^{-3} to 3.77 ng m^{-3} with an annual mean of 1.82 ng m^{-3} for the year 1996. More recently a coordinated study in Europe showed mean TGM concentration in the range of $1.6\text{-}2.4 \text{ ng m}^{-3}$ in the Northern Europe and the Mediterranean area (*Wängberg et al., 2001*).

However *Lee et al.* (1998) mentioned a greater variability for measurements made closer to sources. It is well illustrated by measurements reported by *Poissant (2000)* around Montreal that show low TGM mean concentration with highly variable concentration. Although our mean TGM concentration is obviously higher owing to the proximity of strong Hg sources, our measurements reveal a similar pattern, since the variability and the amplitude of TGM concentrations are important. Nevertheless, we could expect a greater influence of anthropogenic sources with a higher mean TGM concentration. Indeed as we mentioned previously our measurement site is close to a chlor-alkali plant (less than 4 km). Earlier studies conducted near chlor-alkali complex using LIDAR technique (*Ferrara et al., 1992*) or air collection on gold traps (*Maserti and Ferrara, 1991*) exhibited that the mercury levels measured in the atmosphere decrease rapidly a few hundreds meters from the plant. It appears also that background values around $3\text{-}5 \text{ ng m}^{-3}$ are reached in the vicinity (1~2 km) of the plant. Hence, it appears that high TGM values were recorded from time to time reflecting the influence of local anthropogenic sources. However, as regarding the entire set of data, our measurement site seems not to be heavily polluted in comparison with previous studies in urban areas showing higher mean TGM values (*e.g. mean TGM~10 ng m⁻³, Fukuzaki et al., 1986;*

Kim and Kim, 2000; Tan *et al.*, 2000). Moreover, since the spring campaign was hardly influenced by local anthropogenic sources (see discussion in paragraph 3.2), the mean TGM value of 1.9 ng m^{-3} is in very good agreement with values measured during the MOE/MAMCS campaign performed in 1998-1999 (Wängberg *et al.*, 2001).

As regarding short-term variability, we tried to give a careful interpretation of this data. First, due to the limitation of our seasonal database and because of the strong variability of these data due to the influence of local anthropogenic sources, it is difficult to address a precise analysis. Then, trends presented here may reflect the temporal variations of meteorological parameters and also temporal variation in emissions. Indeed there are periods clearly identified (see paragraph 3.2) when an exceptional windy period is affecting significantly TGM data. Concerning the temporal variations in emissions, we established that the main sources around this site were the waste incinerator and the chlor-alkali plant. The other sources were considered as minor sources. Then, sources suspected to be highly variable in emission (such as heating for residential facilities) were supposed not to influence the short-term trends as a result of their minor importance in emission. According to the activity of the chlor-alkali plant and the waste incinerator, we were convinced that their productions remain constant during periods longer than one day. However, a diurnal variation of emission from the chlor-alkali plant may happen and will be discuss after.

According to the mean seasonal TGM concentration, TGM is significantly higher in fall than respectively in summer and winter whereas the mean concentration is lower in spring. Many authors reported various pattern in TGM concentration seasonal variation. Higher concentrations were often observed in winter (Lindqvist, 1991; Kim and Kim, 2000; Poissant, 2000). Our data do not show such a trend suggesting that heating for residential facilities may not represent a significant source of mercury in Champ sur Drac. Influence of such a source may also be hidden by another stronger anthropogenic contribution. Nevertheless, as our data are recorded on about 10

days campaigns in each season, they cannot be significant as a real seasonal trend. Particular meteorological condition and especially atmospheric vertical distribution may influence them.

To elucidate any diurnal pattern within the data, we divided the data both seasonally and diurnally. The diurnal variability is displayed on a ratio scale, calculated as deviation from the mean concentration in figure 4. We are aware of the difficulty to reveal a clear diurnal pattern for a site under obvious anthropogenic disturbance. However some phenomenon may be worthwhile to be discuss. In fall, winter and summer, an increase of TGM with the sunset seems to occur. We noticed that in this particular case TGM correlated better with the increase of temperature than with solar irradiation. This fact is clearly illustrated in summer when the TGM concentration increases rapidly concurrently with the first hot hours of the day. We speculated that it could be an illustration of both emission and reemission processes. On one hand it could reflect the volatility of many Hg species (mostly elemental mercury) emitted from natural areas around our site (Lindberg *et al.*, 1991). On the other hand reemission of Hg species previously deposited is likely to occur. Therefore, temperature and solar irradiation could be two parameters affecting these processes. An increasing temperature and solar irradiation could activate and dissociate divalent mercury complexes through photoreduction reactions (Lindqvist, 1991). Then, emission processes could be accelerated as the temperature increases. However this is not consistent with midday measurements that should show highest values. Undoubtedly, emission and reemission processes provide a part of the TGM signal, however a clear identification is not achievable owing to the proximity of anthropogenic sources.

We sometimes observed high Hg concentration during the night time. It may represent an influence of nocturnal boundary layers form (Lee et al., 1998), trapping TGM near the surface. As focusing more in detail on night time data, during these events low wind speed and declining air temperature were observed. As Schmolke (1999) suggests, TGM concentration may increase as a result of low air-mass exchange and mixing. Thus it may indicate a local source for atmospheric mercury. Therefore, we cannot exclude the hypothesis of higher mercury emission from the chlor-alkali plant during the night. Because of the lower cost of electricity during the night, the chlor-

alkali plant may increase its chlorine production and in this way the amount of Hg released. A previous study performed in a European chlor-alkali complex has also pointed out this phenomenon (Ferrara et al., 1992). However, this interpretation of seasonal and diurnal TGM variability may be misleading as the hourly mean have been calculated for a modest number of values (roughly 40 values for each hourly mean). Therefore, as a result of the high amplitude of TGM values on a 10-days period, the confidence interval for mean is sometimes too large in order to deal with a significant interpretation.

3.2 Influence of anthropogenic sources

Primary, we attempted to give an assessment of potential mercury sources in the area of Champ/Drac. First, we had to deal with the chlor-alkali plant located in the vicinity of our measurement site. 155,000 t of chlorine are produced yearly in this plant. According to several authors, mercury emission factors for chlor-alkali plants are in the range 1.4-5.3 g t⁻¹ of chlorine (Pacyna and Münch, 1991; Ferrara *et al.*, 1992; US EPA, 1997; Mukherjee *et al.*, 2000). Adopting an emission factor of ~4 g t⁻¹, reported by Ferrara *et al.* (1992) for a similar plant, this chlor-alkali plant may release about ~620 kg y⁻¹ of Hg into the atmosphere. Taking into account emission factors available in the literature and mentioned above, the range of emission would be 220-820 kg y⁻¹. Concerning the municipal waste incinerator in Grenoble, we calculated that ~280 kg of Hg may be injected per year in the atmosphere. This estimation is based on an emission factor of 2 g t⁻¹ of waste burned (Bouscaren and Houllier, 1986) and a total mass of 140,000 t of waste burned in 1999. As the chemical composition of municipal refuse and the emission control technique vary from an incinerator to another, it is difficult to adopt a global emission factor. We have chosen an emission factor, which is thought to correspond better to this incinerator. This emission factor is in agreement with those reported in the literature, *i.e.* in the range 0.3-9 g t⁻¹ (Pacyna and Münch, 1991; Hester, 1994; Pirrone *et al.*, 1996; US EPA, 1997) leading to a wide range of emission between 40 and

1,260 kg y⁻¹. Nevertheless, mercury emissions are probably underestimated while medical wastes are also burned in this incinerator. Indeed the Hg concentrations in medical wastes may be 10 to 50 fold higher (Pirrone et al., 1996) than that for municipal wastes. The heating company of Grenoble uses coal, oil, natural gas, wastes and wood as fuels. We estimated that this boiler may emit ~11 kg y⁻¹ of mercury. We had access to information concerning the crematorium of Grenoble. With an emission factor of ~3 g/body (OSPAR, 2000), its contribution was found to be approximately 6 kg per year.

In order to examine the influence of anthropogenic sources on our measurement site, we sorted in figure 5 TGM concentration above 10 ng m⁻³ related to wind direction. We have chosen to keep wind direction values that wind vector component should be at least 1 m s⁻¹. Most of the TGM peaks occurred under north-northwest or north wind condition. This wind direction corresponds to air masses travelling over industries located at the north of our site and also over the city of Grenoble. Thus mercury is probably generated by local point sources in Champ sur Drac such as the chlor-alkali plant and others chemical industries using fuel combustion and also by Grenoble sources included waste incinerator and heating company.

As discussed earlier the TGM concentration was lower in spring than in all other season. It appears clearly that the spring monitoring period was quite unusual (Fig. 6). We identified 3 windy periods (period 1,2 and 3, see Fig 6) lasting approximately one day. As a result of a continuous wind, the accumulation of trace gases emitted by local sources as SO₂, NO and TGM in the atmosphere was less efficient. Between these periods the wind speed was weak and we recorded high TGM values attributed to local anthropogenic sources. Indeed during the windy periods, the site was exposed to air masses coming mainly from the South. This area is a mountainous region where no important sources of mercury were identified. Consequently air parcels from the south are thought to be clean in mercury. Hence, the impact of local anthropogenic sources seems to be a factor affecting significantly TGM concentration in the air of Champ sur Drac.

As we focused more in details on TGM and SO₂ data, we observed the existence of similar variation between TGM and SO₂ concentration. These similarities between TGM and SO₂ peaks are especially well illustrated during the summer campaign (Fig. 7). The correlation coefficient for this period is $R^2 = 0.35$ (N = 150). It might suggest that some fuel combustions contribute to TGM signal. However this relationship is weak and combustion sources cannot be discriminated because there are events when TGM follows the concentration of SO₂ quite similarly in time, but it also exists SO₂ concentration peaks without corresponding increase of TGM. Combustions around Champ sur Drac may provide a part of atmospheric mercury signal. Nevertheless, wind direction dependence of some high TGM values and occurrence of TGM peaks that cannot be explained by closest combustion sources, indicate a mixture of pollution from the chlor alkali plant and the waste incinerator.

4. Conclusions

The high variability of TGM values is the first clear evidence of the influence of local anthropogenic sources. We estimated that the main local anthropogenic sources were the chlor-alkali plant and the waste incinerator (we speculated a total amount of ~900 kg Hg emitted per year). A similarity between SO₂ and TGM concentrations profiles was sometimes observed that may indicate common sources.

Relationship between emission sources north of the site and wind direction was clearly identified. Most of high TGM values were observed concurrently with a north wind. Moreover during the spring measurement campaign, when the wind was stronger and/or from the south (where no Hg sources were identified), mean TGM concentration reached background values of 1.9 ng m⁻³.

For the first time, an extended TGM database has been constituted in France. Since these data are scarce in France, and in order to develop for instance atmospheric transport models in Europe, such database are clearly needed.

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Figure captions

Figure 1: Map of Grenoble area located in the south east of France.

Figure 2: Sampling site location.

Figure 3: Temporal variability of TGM (15 minutes data) at Champ sur Drac in 1999-2000.

Figure 4: Diurnal variability of TGM, (a) in fall, (b) in winter, (c) in spring, (d) in summer, presented on a ratio scale, calculated as deviation from the mean.

Figure 5: Gaseous mercury concentration vs. wind direction. Concentration scale starts from 10 ng m⁻³.

Figure 6: TGM concentration and wind speed measured from 10 to 20 April 2000 (Spring campaign)

Figure 7: Variations of TGM and SO₂ concentrations during summer 2000 (hourly mean).

Table 1: Statistical summary of TGM data at Champ sur Drac in 1999-2000.

Season	Period	TGM (ng m ⁻³)						
		Mean	S.D.	Min.	25% percentile	Median	75% percentile	Max.
Fall	11/04/99-11/16/99	4,8	4,0	0,5	2,1	3,1	6,3	26,9
Winter	01/12/00-01/24/00	3,1	2,4	0,1	1,3	2,8	4,6	16,1
Spring	04/10/00-04/20/00	1,9	2,3	0,2	0,6	1,0	2,3	17,3
Summer	07/17/00-07/31/00	3,7	4,7	0,1	1,0	2,1	4,5	37,1
Overall		3,4	3,6	0,1	1,1	2,3	4,4	37,1

FIGURE 1

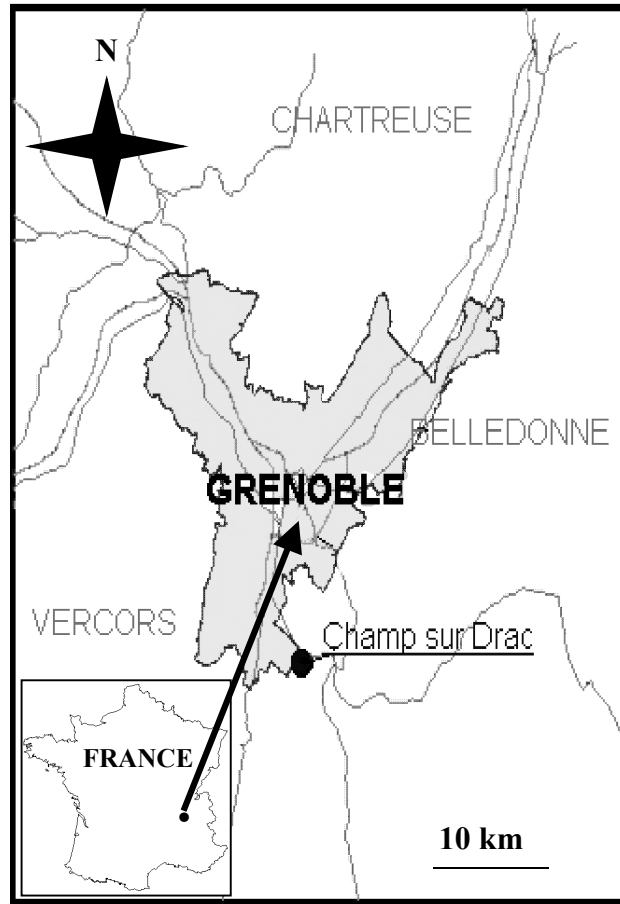


FIGURE 2

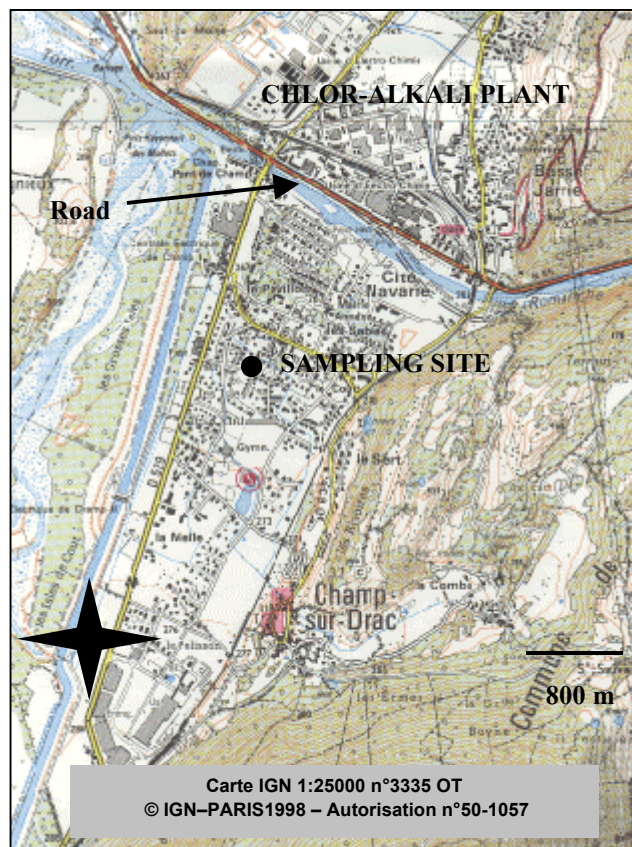
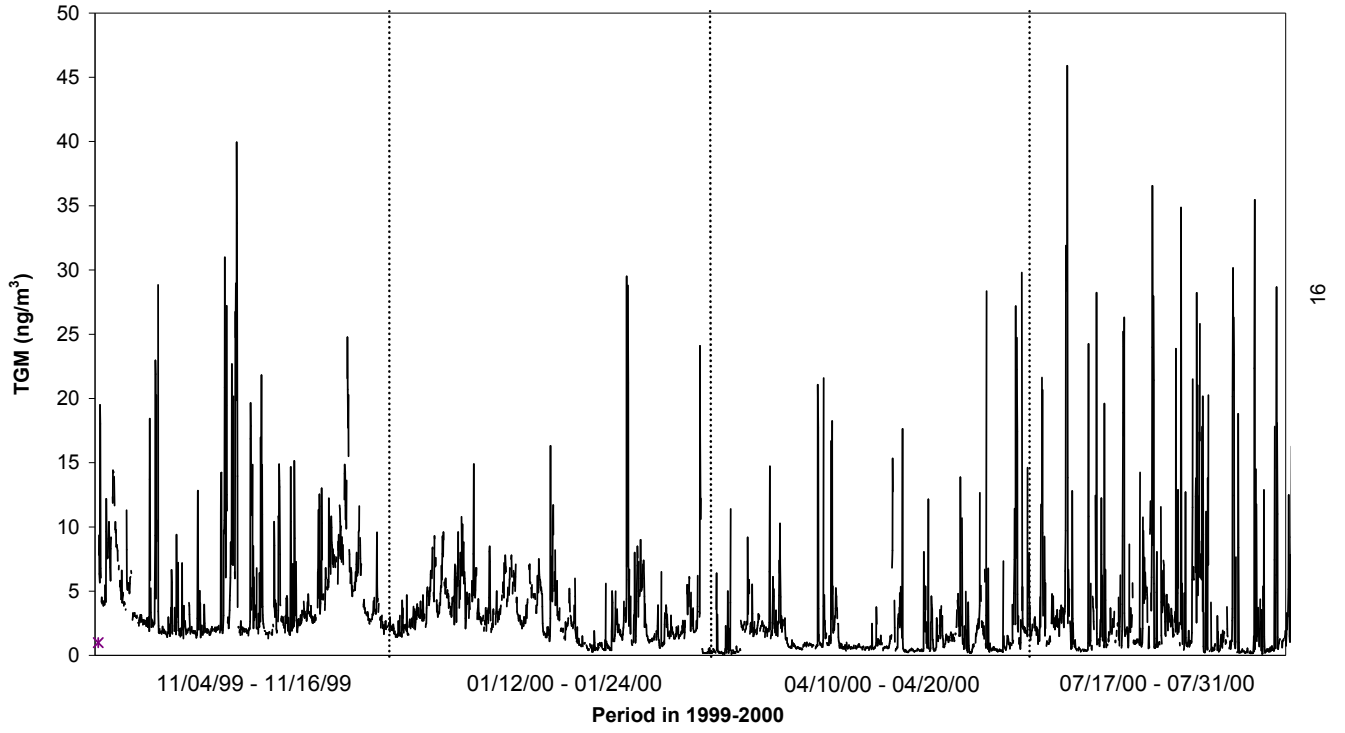


FIGURE 3



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FIGURE 4

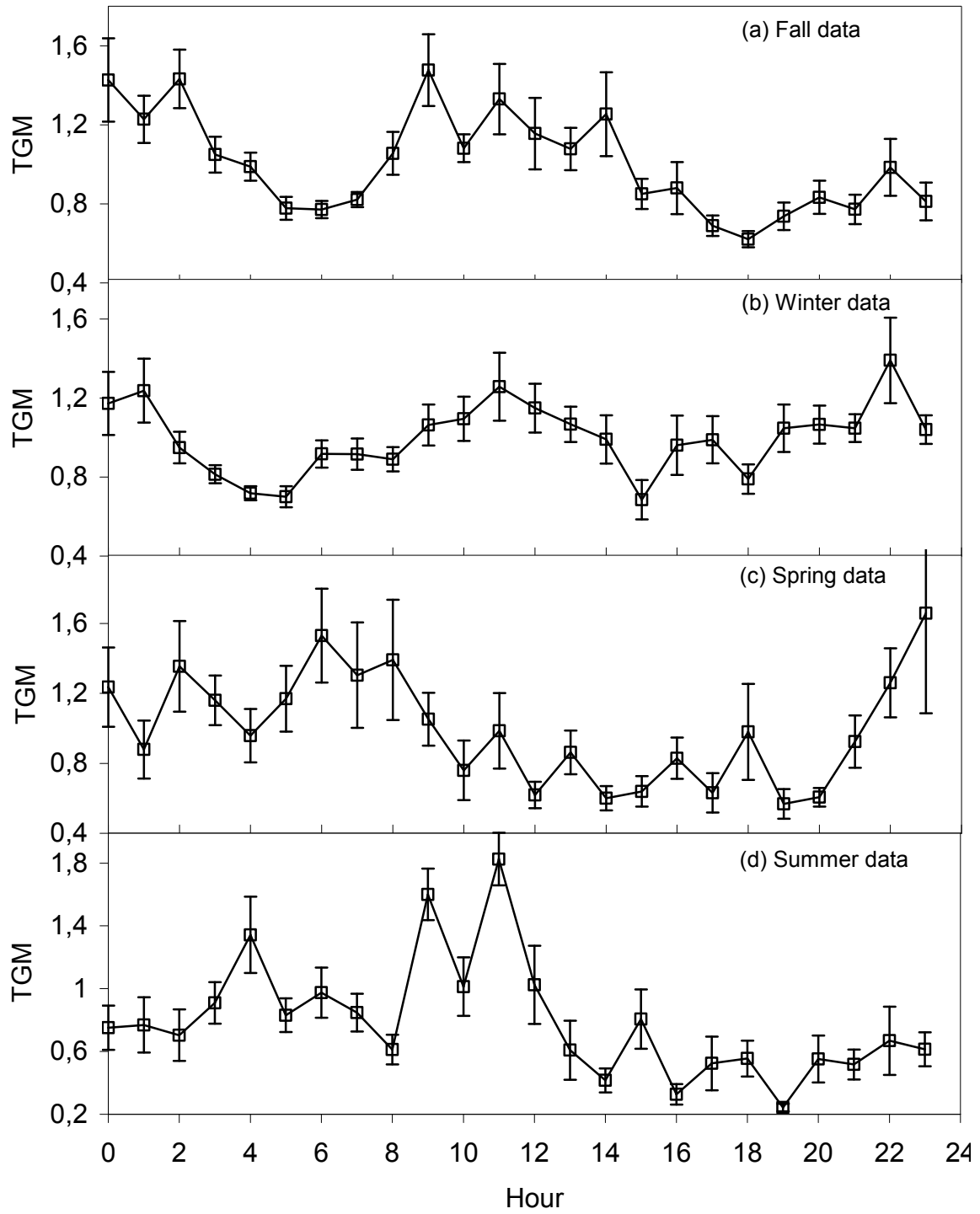


FIGURE 5

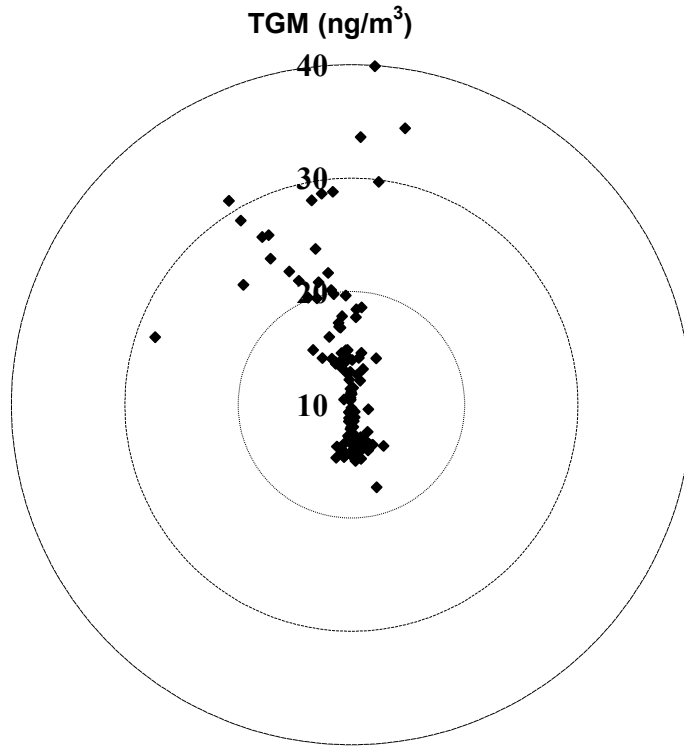
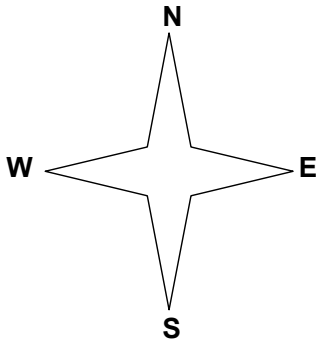


FIGURE 6

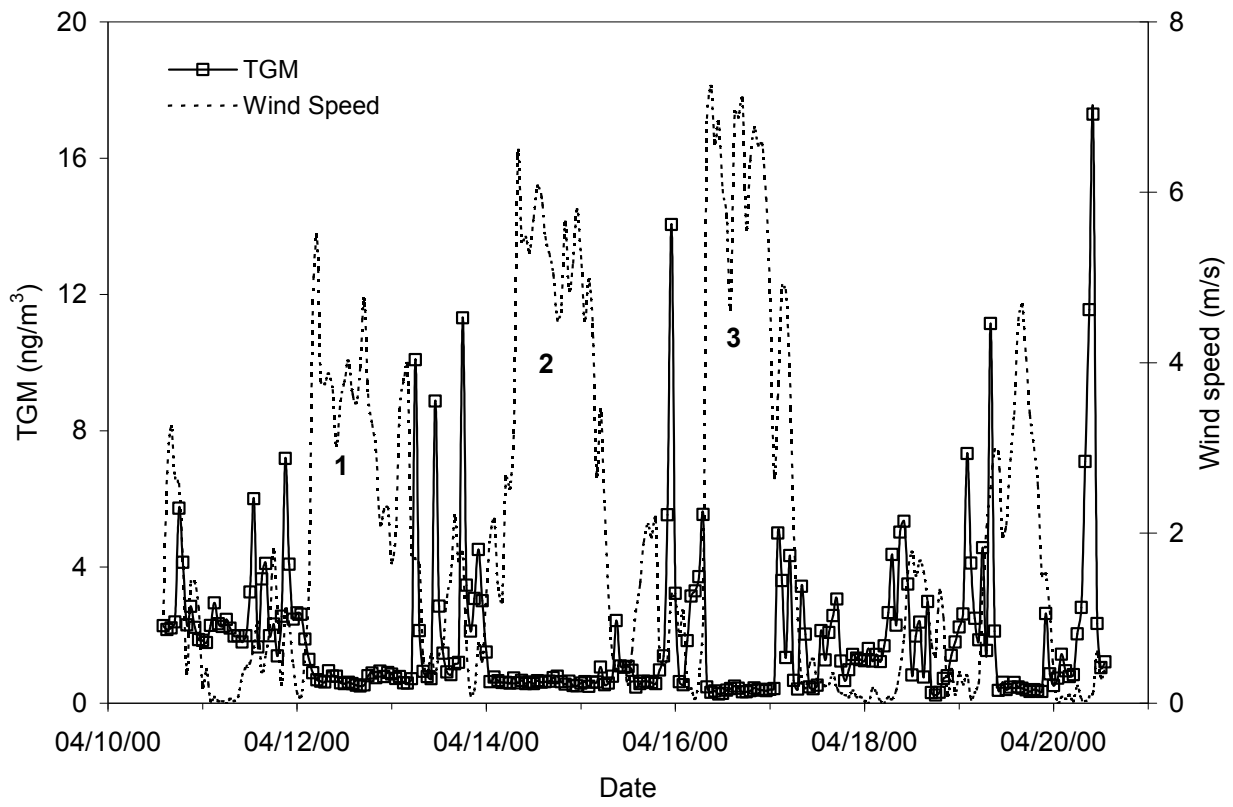


Figure 7

