MASS TRANSFER MEASURMENTS USING A TRACER GAS

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Abstract

The use of artificial tracer gas in natural or industrial configurations made possible to validate diffusion model calculation. Recently applied on volcanoes it gave an accurate evaluation of the volcanic atmosphere pollution and made possible to draw a risk map in case of a toxic gases diffusion in the atmosphere.

Introduction:

Emergency management on atmosphere contamination in natural or industrial configurations presents some similarities:
In both cases, the location of the emergency is well-known, but the time of its occurence is not easy to predict.
The prediction of the area affected and the determination of the concentration reached can be calculated, but it is very difficult to validate these models, taking into account the interaction between the air circulation and the specificity of the local orography.
During the last decade, we developed the use of artificial gas tracing, in order to validate the modelings: this method was used to quantify mass transfers, simulate an emergency situation , and optimise the location of installations or instruments.

Volcanic fluxes determination

The fluxes of volcanic gas and aerosols including HCl and HF are worldwide estimates in using a remote measurement of SO_2 by correlation spectrometry. SO2 flowrates are deduced from SO2 concentrations measured in the plume, plume size and velocities. The flowrate of other species is deduced from the ratio of concentrations measured on samples taken in the cold plume and their ratio to SO2.

The accuracy of this method is dependent on the invariability of the SO2 . Source measurements and modeling of the magmatic gases have shown the complex chemistry occuring during the gas to particle conversion occuring during the cooling of magmatic gases in the atmosphere.

It is very important to compare this classic method using a reactive natural gas with a measurement made simultaneously on the same volcano and involving the non-reactive SF6. artificial gas:

Method used:

In the real atmosphere the turbulences quickly homogenise the concentrations. At less than 1 % (10000 vpm), the density of the mixture is almost equal to that of pure air and the differences in density caused by the difference of molecular mass are negligible when compared with the differences in density caused by the thermal inhomogeneity (effect of expansion on density).

It can be reiterated that a study of contamination transfer is, in fact, a study of the air circulation. Air circulation can be determined by studying the transfer of a particular gas, which is specifically injected into the area believed to be contaminated and which is known as a tracer gas.

We selected the sulphur hexafluoride (SF_6) for two reasons:

SF_6 is a gas of anthropogenic origin and is, therefore, absent from the normal atmosphere. It has limited industrial use (filling electric circuit breakers) and few other applications (medical use, replacing nitrogen for functional tests on breathing). In practice, SF_6 is not detected prior to the experiment and its presence is specific to the tests being carried out.

The gas is clear, has no smell, is not toxic and is one of the most chemically stable gases presently known. It is insoluble in water and solvents and is not trapped as other gases might be.

The main advantages of SF6 are the ease, specificity and the sensitivity with which it is detected.

A simple electron capture detector, a small hollow cylinder with a sheet of ^{63}Ni and an electrode, connected to an electronic board to amplify the signal, enables SF_6 to be detected at 10 vpb (10^{-8} dilution ratio). By placing this detector a few tens of centimetres behind a chromatography column, measurements of 0.01 vpb or less can be achieved without a problem.

An SF_6 analyser has become a portable, autonomous, low-cost piece of equipment, not requiring individual attention. Several analysers can, therefore, be used simultaneously (6 to 8 at present) during one test. Simultaneous measurements in real time enable the air circulation to be determined after only a few tests.

• SF_6 tracer is used at a typical concentration of 1 vpb (mixture ratio 10^{-9}), i.e. 6 $mg.m^{-3}$. The useful range extends from 0.01 to 20 vpb (20 vpb, i.e. 0.12 $mg.m^{-3}$, which corresponds with the saturation of the analysers).

Measurement of gas flow emitted by a precise or diffuse source

The mass flow of gas emitted by the source is calculated by the experimental determination of the **C**oefficient of **A**tmospheric **T**ransfer (CAT of a tracer gas).

The coefficient of atmospheric transfer K observed at a point and relative to a given emission under defined meteorological conditions is the relationship between the concentration C of the effluent and the flow Q of this effluent when the flow is constant and the permanent state has been reached.

$$K = C/Q$$

It is expressed in $s.m^{-3}$.

The coefficient K is obviously a function of the meteorological conditions; it is, however, independent of the nature of the emitted effluent. The method consist in determining K by emission of tracer gas under conditions as close as possible to those of the emission and in carrying out simultaneously at the same point measurements of concentration of tracer gas and gas emitted by the source. The flow of the latter is given by:

$$Q_{gaz} = C_{gaz}/K$$

In order to validate these evaluations we used the two different methods on three different volcanoes: Mt Etna (Sicily), Mt Satsuma Iwojima (Japan) and Mt Erebus (Antarctica). On each one, we operated the correlation spectrometry and at the same time the tracer gas method that can be described as follows: a bottle of SF_6 gas was lowered inside the crater, in order to inject the artifical gas into the volcanic plume. 10 to 20 sampling bottles were installed downwind on the crater rim, in order to sample the plume. By analysing the concentration of gas or aerosols collected at the same place at the same time it was possible to determine the volcanic source output, assuming that the diffusion laws are the same for the artificial and natural products.

Results

On Mt Erebus (Antarctica), the results obtained using COSPEC from a distance gave a SO_2 flowrate of 120 to 150 T/day: i.e. an average of 75 T/day of Sulfur. Using the SF_6 method we obtained 50 to 80 T/day of Sulfur, 150 T/day of Cl and 50 to 80 T/day of F., indicating that the COSPEC method is accurate.

On Mt Etna (Italy), a COSPEC measurement gave a total flux of 4000 to 5000 T/day . The SF6 experiments were processed on Bocca Nuova giving 300 to 400 T/day, i. e., one order of magnitude less than the total flux.

On Mt Satsuma Iwojima (Japan), the SF6 method gave 620 T/day of SO2 whilst COSPEC gave 430 to 900 T/day. The flux of metals was in tons per day: Al: 1.2 to 1.5; Fe: 0.6 to 1.2, Zn 0.15 to 0.22, F: 900T/d anf Cl: 135T/d.

This method was also used to simulate the emergency occuring in case of a volcanic crisis on the "La Soufriere" volcano (French West Indies), last erupting in 1976. Surrounded by a population of 1 000 000 . During the last crisis, we faced the problem of evacuation due to toxic gases. In order to plan the civil defence risk map, we used a SF6 simulation. The tracer gas was emitted from the southern flank of the dome and the concentration measurements were carried out on six portable stations on board small fast moving cars. The complete network was connected by radio. The results were compared with the ash falls observed during the real volcanic crisis and showed a very good correlation: in the trend wind regime, the diffusion of toxic gases takes place along two preferencial channels. The gases flow down slopes from the top of the volcano and rise in altitude when reaching the coast.

Previously, this method was used in urban or industrial systems for the following applications: