

## Forest Soils as Sinks for Atmospheric Gases

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### I. Introduction:

Our understanding of the capacity of various soils to function as sinks for gaseous air contaminants is very limited. This general topic has received only meager research attention over the last twenty years. Obviously, soils have considerable capacity to absorb a variety of gases from the atmosphere, and to incorporate and transform them in or on the soil through a large number of microbial, other biological, physical, and chemical, processes. Specific information is lacking, however, on the relative importance of source versus sink function, the capacities and rates of various soils for absorption, residence, and reaction rate times, the influence of soil physical (mineral and organic matter content, structure, and porosity) and chemical (pH, moisture content, and exchange capacity) properties, climate on removal rates, and significance of soil management practices on removal rates.

Bohn(1972) reviewed the literature on soil sink function and presented some generalizations. He concluded that soils will absorb organic gases more rapidly and in larger quantities with increasing molecular weight and with greater numbers of nitrogen, phosphorus, oxygen, sulphur, and other functional group substitutions in the compound. Absorption of low molecular weight and less substituted organic gases was judged to be dependent on the development of an appropriate microbial population. Soil removal of inorganic gases was concluded to involve primarily chemical and physical processes. The author observed that the literature regarding soil removal of reducing gases was modest, while the information regarding oxidizing gases (ozone, peroxy compounds, and chlorine) was non-existent.

### Carbon monoxide

Carbon monoxide is formed in all combustion processes as a result of the incomplete oxidation of carbon: as a result, anthropogenic production is locally and regionally enormous. In excess of  $6 \times 10^{14}$  g of carbon monoxide is annually discharged into the world's atmosphere (Seiler, 1974). The primary contributors of combustion carbon monoxide are the United States, Europe, and Japan. As a result, most of the anthropogenic emissions are concentrated in the temperate latitudes of the northern hemisphere. Consequently, a key feature of the global distribution of carbon monoxide is a higher concentration in the Northern than in the southern Hemisphere.

Atmospheric carbon monoxide contents are at a maximum during winter and spring and at a minimum during the summer. Over the past two decades, winter concentrations have tended to increase while summer levels have remained constant. Despite the geographic and seasonal variations, the available information supports the conclusion that global carbon monoxide concentrations have remained relatively constant in clean atmospheres. Available data, further support the conclusion that, during the warm season, atmospheric levels of carbon monoxide are related by intense natural sink function (Nozhevnikova and Yurganov, 1978)

A large number of potential natural sinks have been proposed for carbon monoxide:

1. Absorption by oceans,
2. Oxidation to carbon dioxide by  $\text{OH}^\cdot$  in the troposphere
3. Migration to the stratosphere, followed by photochemical reaction,
4. Reaction with animal hememoprotein,
5. Fixation by higher plants, and
6. Absorption by soil

In the light of available evidence, only the latter two hypotheses can be said to be of general importance. Vegetative oxidation of carbon monoxide to carbon dioxide (Ducet and Rosenberg, 1962) and fixation as serine (Chappelle and Krall, 1961), have been described. Employing  $^{14}\text{CO}$ , Bidwell and Fraser (1972) observed uptake by leaves of bean plants under both light and dark conditions. Numerous other non-tree species were tested for carbon monoxide uptake at low gas concentration in the light. Using their bean plant

data, these authors estimated a summer removal capacity of 12-120 kg per km<sup>2</sup> per day, or globally 3-30X10<sup>8</sup> tons per yr.

With the risks of extrapolation from in vitro work with green house plants to the global scene left aside, judgments concerning the significance of vegetation as a carbon monoxide sink remain difficult to make. Inman and Ingersoll (1971) failed to observe any capability of several plants, including seedlings of Monterey and Knobcone pines (*Pinus* species) and Mimosa to remove carbon monoxide from the atmosphere. Also the role of plants as producers of carbon monoxide (Noxhevnikova and Yurganov, 1978) must be more accurately understood before the role of vegetation as a sink for carbon monoxide can be judged appropriately. While vegetation may play some role in the maintenance of carbon monoxide in the natural atmosphere, soils are concluded to be the most important removal agent.

The first evidence supporting the importance of soil as a sink for carbon monoxide was presented in 1926, and this study, along with many that have followed, indicate that soil microorganism are responsible for the removal.

Inman and Ingersoll (1971) conducted preliminary experiments with a green house potting mixture and found that the test soil could deplete carbon monoxide in an experimental atmosphere (containing 120 ppm {13.8X10<sup>4</sup> µg per m<sup>3</sup>} CO) to near zero within 3h. Treatment of the soil with steam sterilization, antibiotics, salt, and anaerobic conditions, all prevented carbon monoxide uptake and indicated the importance of biological processes. A variety of soils from California, Hawaii, and Florida, were brought into the laboratory and tested for their ability to remove carbon monoxide. Inman and Ingersoll generalized from their results that cultivated soils were less active than natural soil and that higher organic matter and lower pH soils were the most active. These observations clearly support the potential importance of forest soils

In order to improve the confidence of their observations, Inman and others outfitted a mobile laboratory and field tested soils in most major vegetative regions of the United States. Field testing was accomplished by covering a square metre of undisturbed soil and vegetation with a gas-tight chamber. The carbon monoxide uptake rate showed considerable variation in the field, ranging from 7.5 to 109 mg per hr per m<sup>2</sup>. As in the previous work, cultivated soils were invariably of less effectiveness than natural soils.

Ingersoll et al. (1974) concluded that the potential rates of carbon monoxide uptake by the soils of the United States and the world were 505 million and 14.3 billion metric tons per year, respectively. Forest soils are indicated to be of particular significance. If these authors estimate approximate to the natural condition, it must be concluded that temperate and tropical forests play extraordinarily important roles as sinks for global carbon monoxide.

Seiler (1974) has determined carbon monoxide uptake rates for several European soils (location and vegetation unspecified) and has calculated an average flux-rate of 1.5X10<sup>-11</sup> g per cm<sup>2</sup> per sec at 15°C. This rate is very approximately an order of magnitude less than the Ingersoll et al average. Seiler explained the discrepancy by indicating that the latter group had employed initial carbon monoxide concentrations of 100ppm (11.5 X 10<sup>4</sup> µg per m<sup>3</sup>), while his laboratory had employed 0.20 ppm (230µg per m<sup>3</sup>) which was judged to be the normal ambient concentration in unpolluted areas of Europe. Seiler's estimate for global soil removal was 4.5 X 10<sup>8</sup> tons per year. The question of inadequate account of carbon monoxide evolution by soils has been raised by K.A. Smith et al. (1973). This deficiency could contribute to important overestimations of sink function. Perhaps the actual capacity for carbon monoxide removal lies between the estimates of Inman's and Seiler's groups, or perhaps it approximates one or the other.

### **Sulphur Dioxide and Hydrogen Sulphide**

The sulphur containing gases that are currently recognized as important components of tropospheric air include sulphur dioxide (SO<sub>2</sub>), hydrogen sulphide (H<sub>2</sub>S), carbonyl sulphide (COS), carbon disulphide (CS<sub>2</sub>), dimethyl sulphide (CH<sub>3</sub>SCH<sub>3</sub>), and Sulphur hexafluoride (SF<sub>6</sub>) (Brenner and Steele, 1978). Until very recently it was generally assumed that the primary and most important forms of sulphur in the atmosphere were in hydrogen sulphide, sulphur dioxide, and sulphates. The information concerning soils as a sink for atmospheric sulphur has dealt almost exclusively with the latter two. Sulphate is added to soil largely by precipitation, and becomes part of the soluble sulphur content held by soil colloids. Soils also have a large capacity to absorb sulphur dioxide quickly from the atmosphere. Factors that tend to increase the soil uptake of sulphur dioxide include fine texture, high soil organic matter content, high pH, presence of free CaCO<sub>3</sub>, high soil moisture content, and the presence of soil microorganisms (Nyborg, 1978).

Since always et al. (1937) presented the first evidence that soils can absorb sulphur dioxide, a large number of studies have followed. K. A. Smith et al (1973) studied the capacity of six soils with variable chemical and physical properties, from Oregon, Iowa, and Saskatchewan, and found that removal by them of sulphur dioxide and hydrogen sulphide from the atmosphere was much more rapid than the removal of carbon monoxide. It is not clear if any of the soils were from forest ecosystems, but the pH (4.8) and organic carbon percentage (9.38) suggest that the Astoria, Oregon, soil may have been. Clearly soil moisture favors uptake of

sulphur dioxide-presumably owing to the high solubility of this gas in water. The Astoria soil appears quite average in its ability to remove sulphur dioxide and Hydrogen sulphide. Bohn (1972) has suggested that the absorption rate of hydrogen sulphide slows with high soil moisture contents, perhaps due to slow diffusion rates in water filled pores.

The fact that hydrogen sulphide absorption capacity increases with higher soil pH, may reduce the importance of forest ecosystems in removing this gas from the atmosphere. Despite the relatively large number of papers addressing the soil removal of this gas and sulphur dioxide (e.g. Moss, 1975; Bremner and Steele, 1978), estimates of regional and global removal amounts by soil are relatively few. Available data suggest that deposition velocities ( $v$ ) for sulphur dioxide are generally in the range of 0.2 to 0.7 cm per sec (Rasmussen et al., 1974) which is less than those for vegetation. Abeles et al. (1971) examined the capacity of soil collected from Waltham Massachusetts, to remove sulphur dioxide under laboratory conditions (100ppm { $26.2 \times 10^4 \mu\text{g per m}^3$ } gas) and extrapolated their results to suggest that United States soils may be capable of removing  $4 \times 10^{13}$  kg of sulphur dioxide ( $2 \times 10^{13}$  kg sulphur dioxide-sulphur) per year.

The gas chromatographic studies of Bremner and Banwart (1976) showed that air dry and moist soils had the capacity to absorb dimethyl sulphide, dimethyl disulphide, carbonyl sulphide, and carbon disulphide, but did not absorb sulphur hexafluoride. The first four gases were removed more efficiently by moist soils than dry ones. Soil sterilization indicated that soil microorganisms were partially responsible for the removal of these gases. As the rates of removal were substantially less than those for sulphur dioxide or hydrogen sulphide, however, the Authors concluded that, while soils may constitute a sink for low levels of dimethyl sulphide, dimethyl disulphide, carbonyl sulphide, and carbon disulphide, they would not effectively reduce elevated levels of these gases in areas of high anthropogenic emission.

### **Nitrogen oxides and other nitrogen-containing Gases**

In the atmosphere, nitric oxide is either oxidized to nitrogen dioxide or photolysed to nitrogen gas. Nitrogen dioxide reacts photochemically or is removed by precipitation, primarily in the form of nitric acid. Nitric oxide and nitrogen dioxide may also be removed by soils. Nitric oxide is oxidized to nitrogen dioxide in soil, but the former gas does not persist in acid soils as long as it does in basic soils (Bohn, 1972).

Working with Waltham, Massachusetts, soil, Abeles et al. (1971) found that the removal rate for nitrogen dioxide was slower than the removal rate for sulphur dioxide. Twenty four hours were required to reduce nitrogen dioxide concentrations from 100 to 3ppm ( $18.8 \times 10^4$  to  $56.4 \times 10^2 \mu\text{g per m}^3$ ) in test atmospheres. Extrapolation of these laboratory experiments allowed these authors to suggest that the soils of the United States may be capable of removing  $6 \times 10^{11}$  kg of nitrogen dioxide per year

Both Ghiorso and Alexander (1976) and E. A. Smith and Mayfield (1978) have documented rapid absorption of nitrogen dioxide by both sterile and non sterile soil. In the latter study, soil from uncultivated grassland in Ontario absorbed 99% of the nitrogen dioxide introduced into a test vessel at  $25^\circ\text{C}$  in 15min.

Mechanisms of nitrogen dioxide uptake may involve reaction with soil cations to form  $\text{NaNO}_2$  or  $\text{KNO}_3$  reaction with soil water to form  $\text{HNO}_2$  and  $\text{HNO}_3$ , binding with organic matter, or persistence as a gas in inter particle soil spaces (E.A. Smith and Mayfield, 1978).

As ammonia would probably be present in the atmosphere in the form of  $(\text{NH}_4)_2 \text{SO}_4$  rather than  $\text{NH}_3$  in all but the most unusual environments, direct soil absorption of gaseous  $\text{NH}_3$  is probably not important. Where ambient conditions might expose soils to high ammonia levels, however, evidence suggests that acidic soils are particularly efficient removal agents. The specific capabilities that forest soils may have to remove nitrogenous gases must await further experimentation.

### **Hydrocarbons:**

Hydrocarbons are generally not soluble in water, and as a result, soil uptake where it is important, is concluded to be primarily microbial. The light hydrocarbon from motor vehicles that is most actively removed soil, is ethylene (Zimmerman and Rasmussen, 1975). Abeles et al. (1971) observed that Maryland soil samples removed ethylene more slowly than other soils removed sulphur and nitrogen dioxides. Soil removal of ethylene, mediated by various microorganisms, was calculated by Abeles's group to approximate  $7 \times 10^9$  kg of ethylene annually in the United States.

K.A. Smith et al. (1973) determined that the soil flux-rate for acetylene was from 0.24 to 3.12 per 10 mole per g per day. Their test soil with the lowest pH and highest organic matter (forest soil) was the most active of all soils tested for acetylene removal.

### **Oxidants:**

There is limited evidence that soils function as a sink for atmospheric ozone (Rasmussen et al., 1974).

Aldaz (1969) concluded that the soil and vegetation of the surface of the Earth represent a major sink for this gas, and estimated the capacity of this sink to be within the range of 1.3 to  $2.1 \times 10^{12}$  kg ozone per yr.

Most reports of ozone removal have examined plant uptake or plant and soil uptake combined. Turner et al. (1973) have tested the sink capacity of a freshly cultivated sandy loam devoid of vegetation. Their results, which were recorded under field conditions, showed that the flux rate of ozone removal varied from 3 to  $12 \times 10^{11}$  mole per  $\text{cm}^2$  per sec, making bare soil in these authors judgment an important sink for ozone.

#### Other Gases:

Fang (1978) examined the uptake of mercury vapour by five Montana soils by exposing them to a test atmosphere containing  $75.9 \mu\text{g}$  metallic  $^{203}\text{Hg}$  vapour per  $\text{m}^3$  for 24h. The soil with the highest organic matter content showed the highest mercury uptake. While mercury vapour is currently only an extremely localized problem, more than 90% of the mercury contained in coal is vaporized during combustion. Even with relatively low mercury concentrations in coal, widespread or large volume coal combustion may increase the significance of soil retention of mercury vapour.

## II. Summary

Forest soils are important sinks for a variety of air contaminants. Retention of particulate lead by organic materials in the forest floor is most dramatic example. The flux of lead to temperate forest ecosystems downwind of industrial, urban, or roadside, sources may approximate 200-400g per ha per yr, with much of this lead accumulating in the forest floor. Certain forest soils may also serve as a sink for additional trace metals, including zinc, cadmium, copper, nickel, manganese, vanadium and chromium. The efficiency of sink functioning for the latter metals is generally substantially less than for lead, but may be important-particularly in forest systems close to primary sources.

Forest soils remove pollutant gases from the atmosphere via several microbial, chemical, and physical, processes. Forest soils function as an especially efficient sink for carbon monoxide, and may play a dominant role in regulating the concentration of this gas in the atmosphere. Other gases that may be significantly removed by forest soils include sulphur dioxide, ammonia, some hydrocarbons, and mercury vapour. It can be concluded that forest soils play a role of very significant importance as repository for atmospheric carbon monoxide. As the soil removal rate increases with increasing levels of carbon monoxide, forest ecosystems in and around urban and industrial areas may be especially important sinks.

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