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Spatial and temporal distributions of mercury emissions from forest fires in Mediterranean region and Russian federation

Sergio Cinnirella*, Nicola Pirrone

CNR-Institute for Atmospheric Pollution, Division of Rende, c/o UNICAL, 87036 Rende, Italy

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Abstract

Mercury emissions from forest fires in Europe and in North African countries bordering the Mediterranean Sea were estimated on the basis of ground-based forest fires data, forests phytomass and measured emission factors. Satellite observations were used to assess mercury emissions in Russian Federation through the assessment of spatial and temporal distributions of forest phytomass content and forest burnt areas. Annual mercury emissions in Europe were in the range of 0.9–3.6 $Mg\,yr^{-1}$, Italy and Portugal have the highest emissions with an average of 821 and 459 $kg\,yr^{-1}$ respectively, followed by Spain (394 $kg\,yr^{-1}$), France (247 $kg\,yr^{-1}$) and Poland (191 $kg\,yr^{-1}$). On regional scale, Hg emissions from this particular source in the Mediterranean region is 2.3 $Mg\,yr^{-1}$ (in the range 0.4–4.0 $Mg\,yr^{-1}$) on average, while emissions from African and Middle East countries bordering the Mediterranean Sea are not significant. Mercury emissions in the Russian Federation are in the range of 3.4–24.8 $Mg\,yr^{-1}$ (16 $Mg\,yr^{-1}$ on average).
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1. Introduction

Highly resolved spatial and temporal distributions of mercury emissions to the atmosphere from natural and anthropogenic sources are needed in global mass balance models, transport and deposition models, for relating mesoscale variations in mercury concentrations in both gas and particulate phase with regional and global circulation patterns, and in assessing the long-term ecological and health impacts on different environmental compartments (Pirrone et al., 1996; Pacyna et al., 2001; Pirrone et al., 2001, 2003).

It is well known that mercury is released to the atmosphere from a multitude of anthropogenic and natural sources (Pirrone et al., 1996; Pacyna et al., 2001; Pirrone et al., 2003). The ratio between the relative contributions of these source categories may vary with region and time of the year. On global scale the contribution from industrial sources ranges between 1660 and 2200 $t\,yr^{-1}$ (Pirrone et al., 1996; Pacyna et al., 2001, 2003), whereas emissions from natural sources (i.e., volatilization from water surfaces, volcanoes, re-emissions from topsoil and vegetation) may represent the major contribution (up to 60% of the total) to the global atmospheric mercury budget (Pirrone et al., 1996,

*Corresponding author. Tel.: +39 0984493206 (direct); fax: +39 0984493215.
E-mail address: s.cinnirella@cs.iia.cnr.it (S. Cinnirella).

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2001). In Europe mercury emissions from natural sources account for about 40% of the regional total (Pirrone et al., 2001, 2003); however, this percentage may be much higher (up to 60% of the regional total) in the Mediterranean region (Pirrone et al., 2001, 2003). Among natural sources the emission from volcanoes, forest fires and surface waters represent a significant contribution and also emissions from contaminated soils in ancient mining industrial areas or particular geologic units rich in Hg (i.e. capgaronnite, cinnabar, cordeorite) can also be significant (Ferrara et al., 1998; Gustin et al., 2002).

In recent years the evaluation of mercury emissions from phytomass burning has received a growing attention primarily due to its potential significant contribution to the regional atmospheric mercury budget (Veiga et al., 1994; Carvalho et al., 1998; Roulet et al., 1999; Friedli et al., 2001, 2003; Sigler et al., 2003; Pirrone et al., 2005).

Mercury in vegetation originates from several mechanisms, including the uptake from the atmosphere, atmospheric deposition to foliage and uptake from roots (Rea et al., 2002); however, the proximity of vegetation to natural or anthropogenic sources (hot spots or contaminated sites) may increase its Hg content (Lodenius, 1998; Carballeira and Fernandez, 2002; Lodenius et al., 2003). Therefore, when a forest burns particulate and gaseous mercury is released to the atmosphere or it is transported by runoff into rivers and/or other water reservoirs (Caldwell et al., 2000; Amirbahman et al., 2004). Mercury release from forest fires has been estimated on the basis of direct or indirect measurements of Hg concentrations in smoke of boreal wildfires (Brunke et al., 2001; Friedli et al., 2001, 2003; Sigler et al., 2003), or tropical forest with intense phytomass burning (Veiga et al., 1994; Artaxo et al., 2000). However, very little has been done to downscale Hg emissions in space and time by using highly resolved spatial data that are useful for regional and global scale atmospheric models.

During the last decade several studies have assessed Hg emissions from forest fires in North and South America as well as in Boreal regions (Veiga et al., 1994; Artaxo et al., 2000; Brunke et al., 2001; Friedli et al., 2001, 2003); however, very little has been done to assess Hg emissions from forest fires in Europe and Mediterranean region.

The aim of this paper is to present a first assessment of spatial distributions and temporal trends of Hg emissions from forest fires in Europe, Mediterranean region and Russian Federation. Emissions in Europe and Mediterranean region were derived from ground-based data, while satellite observations were used to estimate Hg emissions in the Russian Federation.

2. Methodology

Mercury emissions from forest fires (Q_{Hg}) (in kg of mercury per year) can be estimated as the sum of mercury released from vegetation and that from soil. Mercury emission from vegetation burning was estimated as (Roulet et al., 1999):

$$Q_{Hg} = AM\mathrm{Hg}_c E_c E_{Hg}, \quad (1)$$

where A is the burnt area (ha yr^{-1}), M is the amount of standing phytomass (kg ha^{-1}), Hg_c the mercury concentration in vegetation (μg kg^{-1}), E_c the efficiency of phytomass combustion and E_{Hg} is the efficiency of mercury released to the atmosphere.

The mercury released to the atmosphere from vegetation is primarily related to mercury concentrations in foliage (Hg_c). It varies with tissues of plants—predominantly leaves, bark and root-, foliar age, forest typologies as well as soil characteristics (Rasmussen, 1994; Fischer et al., 1995; Lindberg, 1996; Friedli et al., 2001; Ericksen et al., 2003). Hence, in proximity of contaminated sites Hg concentration in vegetation may vary significantly due to the high Hg concentrations in soil and air (Ericksen and Gustin, 2004). Moreover, the foliar age and the species physiology contribute to the final Hg amount in vegetation (Rea et al., 1996; Schwesig and Krebs, 2003), while the phenological cycle of the plant affects its retention because of deciduous plants transfer accumulated mercury to soil through litterfall (Amirbahman et al., 2004). In addition, mercury accumulated on foliage through atmospheric deposition represents an important contribution of Hg loads entering to the soil. The vegetation conditions (i.e. phytomass amount, tissues' water content) may have a significant influence on mercury emissions from phytomass burning; the typology of the forest fire also affects Hg concentration in smokes. In particular, emissions from crown fires are less than that from litter fires because of the highest Hg concentration in litter that may be up to 71 ng g^{-1} (Friedli et al., 2003).

The forest combustion efficiency (E_c) may vary significantly with type of ecosystem and changing climatic and hydrological conditions. The combustion

efficiency may vary between 0.5 and 1.0 depending upon the air/fuel ratio, fuel type, fuel load, fuel chemistry, fuel moisture content, packing ratio for the fuel particles, surface area to volume ratio and method of ignition (Ward et al., 1992, 1996). The efficiency of mercury released to the atmosphere (E_{Hg}) can be up to 97–99% implying a complete transfer of Hg from vegetation to the atmosphere (Friedli et al., 2003). In conclusion, E_{Hg}, E_c and Hg_c can be grouped into a single factor known as Emission Factor which varies from 14 to 71 μg/kg of dry phytomass (Friedli et al., 2003).

In the case of soils, mercury released to the atmosphere is strongly controlled by the Hg substrate concentration, light intensity and temperature (Ferrara et al., 1997; Engle et al., 2001; Zehner and Gustin, 2002). Field measurements suggest that mercury release from the top soil during a forest fire is primarily dependent upon the increase of temperature caused by the activation of complex flux processes from the lower to the upper soil horizons (Iglesias et al., 1997), leading to an Hg flux of 1–5 $mg\,m^{-2}$ (Woodruff et al., 2001). The litter typology can also play an important role in the Hg emission during a forest fire.

Therefore, during a forest fire both vegetation and soil contribute to release Hg to the atmosphere, however the difficulties in assessing the relative emission factors for these two sources suggest to use a global emission factor (E_f). In our assessment the E_f measured in field campaign was used.

Friedli et al. (2003) found that E_f was in the range of 112 ± 17 μg of mercury per kg of fuel and varies with phytomass type and other environmental factors (i.e. ambient temperature, relative humidity, mercury content in vegetation and soil). The mercury species released during a forest fire are primarily elemental gaseous mercury (GEM) and particulate mercury (Hg(p)), while the fraction of reactive gaseous mercury (RGM) is generally negligible (Friedli et al., 2003; Sigler et al., 2003). Hence, Eq. (1) can be re-written as

$$Q_{Hg} = AME_f. \quad (2)$$

Eq. (2) was used to estimate mercury emissions (Q_{Hg}) with ground-based and remote sensing data.

Due to limited information available a common E_f was adopted in assessing the spatial distributions of mercury emissions, whereas the phytomass amount and burnt area were estimated on the basis of ground-based data and RS observations. Our assumption ($E_f = 112 \pm 17\,\mu g\,kg^{-1}$) reflects the average E_f estimated from literature (Fig. 1). On the basis of biomass-mercury release ratios the average E_f for temperate/boreal forests results to be of 111 and 83 $\mu g\,kg^{-1}$ for tropical forests. As reported above, E_f is dependent upon the mass of fuel (Friedli et al., 2003), including the emissions from

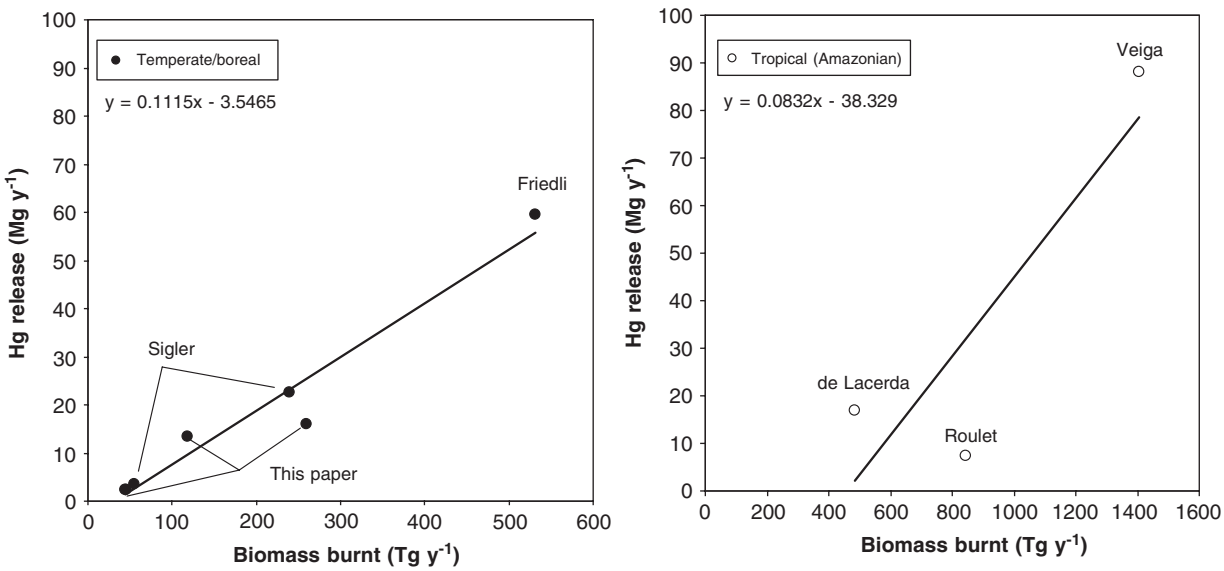

Fig. 1. Biomass-Hg release ratios as estimated by authors reported in Table 4. The angular coefficient of regression lines gives the amount of mercury released per unit of burnt biomass for temperate/boreal (111 $\mu g\,kg^{-1}$) and tropical (83 $\mu g\,kg^{-1}$) forests.

both soil and vegetation as well as different Hg content in vegetation, different combustion efficiency and different emission efficiency, hence the M factor in Eq. (2) represents the standing phytomass.

Keeping the above in mind, the total organic matter in a forest (zoomass and phytomass as well as dead or living mass) represents the biomass, while the aboveground (stems, branches and leaves) and belowground (roots) plant mass represent the phytomass, whereas in the narrow sense the phytomass represents the dry weight of aboveground living plant mass which does not account for the dead phytomass (dead wood and litter). Hereafter the term "phytomass" has been used with reference to the dry aboveground phytomass. The phytomass is the balance between production and consumption and changes related to intrinsic forces (i.e. growth) as well as to external forces (i.e. anthropogenic and natural disturbances) (Brown et al., 1999). In our case the phytomass represents the plant mass before the fire event.

The topsoil is defined as "The upper part of a soil, with the lower limit set at 30 cm or shallower" FAO/UNEP (1999). For convenience it could be selected as the A0–A1 horizon. It changes with the type of ecosystem and climatic conditions.

2.1. Burnt areas and phytomass amount in Europe and Mediterranean region

The occurrence of forest fires in summer and dry winter periods is frequent in the Mediterranean region and represents a growing management and environmental concern in several countries including Italy, Greece, Portugal, Spain, Southern France, African and Asian countries facing the sea. Spatial data on burnt forest areas was derived from ground-based data sets provided by national statistic offices and, in a small number of cases, was integrated with satellite data to get more recent spatially resolved information. In the first case, data are supplied to statistical offices by direct observations of forest agencies that, however, do not provide geographical references. When using data provided by national forest offices the difference related to the definition of what has to be considered as forest land must be taken into account. Forest area assessed in all national surveys and forest statistics (both at national and international level) always include the crown cover percentage and surface size as driving criteria (FAO/UNEP, 1999). Clearcut areas and young stands which do not fulfill the criteria are often included in the forest areas (Päivinen and Köhl, 1996).

In most parts of Europe the impact of discrepancies on the amount of total forested area might be small with the exception of the Mediterranean region where forests are naturally open and show natural gaps. In the Global Forest Resource Assessment (FRA) reports published by FAO (1995, 2000) the following definition of forest land was adopted: "*Land with tree crown cover of more than 10 percent and area of more than 0.5 ha. The trees should be able to reach a minimum height of 5 m at maturity* in situ". Based on the methodology proposed in FRA 2000, the FAO developed a fire database at country scale, reported in the Global Forest Fire Assessment 1990–2000 (FAO, 2001). This database was used in our assessment to evaluate the burnt areas; however, in the case of Portugal, Spain, France, Italy and Greece the database was integrated with the European Forest Fires Information System retrieved from satellite observations (Barbosa et al., 2003).

Also the assessment of the phytomass amount is affected by uncertainty due to different definition of the "forest" because statistics were compiled from country information sources following standard terms and definitions. Volume and phytomass data are available for most of industrialized countries, while for developing countries, the phytomass per hectare was calculated for each national forest type using the volume and wood density data, and by integrating the trees' volume with the phytomass of other above-ground forest components (FAO, 2000).

The continuous phytomass growth does not often correspond to measurement data adopted by statistical offices that provide valuable information on multi-annual basis. Commonly, forest phytomass is estimated from forestry inventories that provide the growing stock volume (generally defined as the under-bark volume of main stem greater than a selected diameter). The phytomass is then calculated by considering the dry weight density of tree species. Percentage of necromass (i.e. leaves, twigs, stems) may vary with the forest phytomass (2–28%) and is taken into account assuming that a 10% of phytomass is necromass (Alexeyev et al., 1995). The phytomass data used in our calculation were retrieved from Garzuglia and Saket (2003).

2.2. Burnt surface and phytomass amount in Russian Federation

To test the RS approach the Russian Federation was selected as case study because forests in Russia account for 22% of the world's total and 43% of the total temperate/boreal forests. Statistics on fires show that up to 1.6 million ha of forest are affected yearly by intense fires, that was estimated to be about 35 million Megagrams of phytomass (Megagrams, $Mg = 10^6$ g) generating $2.19\,kg\,m^{-2}$ of burnt fuel (Conard and Ivanova, 1997). Thus, the magnitude of both boreal forest and burnt areas suggests that Hg emission from forest fires may represent an important contribution to the total Hg emission.

AVHRR satellite data was used to derive the temporal (1996–2002) and spatial distribution of burnt areas in Russia. Data were obtained by Hyer (2004) using an algorithm that allows to calculate the probability of occurrence of a fire from the AVHRR thermal channels, which was enhanced by end-of-season mapping of fire scars. Data on yearly burnt areas, available at the Global Land Cover Facility (GLCF, 2004), were downloaded and re-projected to have a unique reference system.

Mercury emissions were estimated on the basis of the phytomass amount and burnt surface data sets for the years 1996–2002. Digital data sets of phytomass are not available; therefore, phytomass data were derived from stand carbon data. Forest Stand Carbon Map of Russia was derived from the mapping of forest tree stands prepared by Alexeyev et al. (1995) and provides the carbon reservoir in the Russian forest (Stone et al., 2003) by summing the amounts of carbon in the phytomass, in dead phytomass and in topsoil. Through the distribution coefficients, which represent the partition between different plant organs, reported by Alexeyev et al. (1995), the carbon storage in phytomass was calculated. Data represent continuous estimates of forest stand carbon in a spatially explicit form. The transformation from map of carbon to phytomass amount was based on the evaluation of carbon content in the vegetation. The phytomass to carbon conversion (and vice versa) depends upon the intrinsic plant characteristics (i.e. wood density, wood fraction), however, the carbon content in plants was found in the range of 0.4–0.5 kg of carbon per kg of dry phytomass (Alexeyev et al., 1995; Conard and Ivanova, 1997; Lakida et al., 1997; Milne and Brown, 1997; Haripriya, 2000) thus a coefficient of 2.0 was adopted in estimating the primary forest and the understorey vegetation phytomass for the Russian region.

3. Results and discussion

3.1. Mercury emissions in Europe and in the Mediterranean region

Forest fires occur mainly in the European Countries bordering the Mediterranean Sea (Cyprus, France, Greece, Italy, Portugal and Spain) with a Mean Annual Burnt Surface (MABS) of ~76,236 ha (Table 1). From 1990 to 2003 the MABS was highest in Spain with 1466 km^2 followed by Portugal (1305 km^2), Italy (1049 km^2), Greece(500 km^2) and France(240 km^2). In contrast, the lack of data for North African countries does not allow any reliable assessment of burnt areas in these countries. The former USSR countries show the

Table 1
Trends of total burnt area (km^2) derived from statistical offices within the European (EU-25) with Norway and Switzerland included, Mediterranean (MED), Euro-Asiatic (EUR-ASIA) and African (AFR) Countries for the years 1990–2003

Year	EU25 + 2[a]	MED[b]	EUR-ASIA[c]	AFR[d]
1990	6520	6615	16,890	36
1991	5601	5768	11,680	54
1992	4177	3961	12,301	40
1993	4196	4654	12,934	45
1994	7485	7790	7982	75
1995	4157	4336	5191	85
1996	2624	2810	24,976	26
1997	3351	3452	13,624	53
1998	6477	6432	54,334	33
1999	3294	3094	10,389	31
2000	7426	8189	22,760	15
2001	3820	3857	16,041	—
2002	2127	2129	—	—
2003	6866	6838	—	—

[a]Countries: Austria, Belgium, Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Netherlands, Norway, Poland, Portugal, Slovakia, Slovenia, Spain, Sweden, Switzerland, United Kingdom.

[b]Countries: Albania, Algeria, Bosnia & Herzegovina, Croatia, Cyprus, Egypt, France, Greece, Israel, Italy, Lebanon, Libya, Macedonia, Morocco, Portugal, Slovenia, Spain, Tunisia, Turkey, Yugoslavia.

[c]Countries: Albania, Armenia, Azerbaijan, Belarus, Bosnia & Herzegovina, Bulgaria, Croatia, Faeroe Islands, Georgia, Gibraltar, Iceland, Israel, Kazakhstan, Lebanon, Macedonia, Moldova, Romania, Russian Federation, Tajikistan, Turkey, Turkmenistan, Ukraine, Yugoslavia.

[d]Countries: Libya, Morocco, Tunisia.

highest value, with a MABS of 1.58 million hectares per year in the Russian Federation. Grouping countries by region, EUR-ASIA accounts for 17,425 km^2 of the annual burnt surface followed by EU-25 with 4606 km^2 and Africa with 2108 km^2, on average.

The phytomass amount depend on the forest typology (i.e. conifer or hardwood, stand or coppice, close or open forest), climate (i.e. Boreal, Continental, Mediterranean), forest species (i.e. pines, oaks, eucalypts) and silvicultural system. Fig. 2 provides the phytomass distribution within the European, Asian and African countries for the year 2000 and highlight the influence of the latitude on the biomass amount suggesting that the same species (i.e. Fagus sylvatica, Pinus nigra) may have a different stocking in a stand in Northern Europe compared to the Mediterranean region. For example, forests in Finland and Sweden have less phytomass per unit area than in Belgium, France or Germany. But several exceptions arise from the figure as consequence of forest typology and forest management system (i.e. the highest values of Austria, Slovenia, Slovak Republic and Switzerland due to the silvicultural tradition in this region). Therefore, the phytomass amount was highest in Austria with 250 Mg ha^{-1} and lowest in Turkmenistan with 3 Mg ha^{-1} with no significant forest areas in Faeroe Islands, Gibraltar, Monaco, San Marino and Vatican City, whereas for several countries including Andorra, Bosnia and Herzegovina, Kyrgyzstan, Luxembourg, Macedonia, Ukraine and Uzbekistan no reliable data was found.

Although data on burnt surface and phytomass consistency are incomplete, and despite the phytomass estimate reveals the limits of a mean value, a

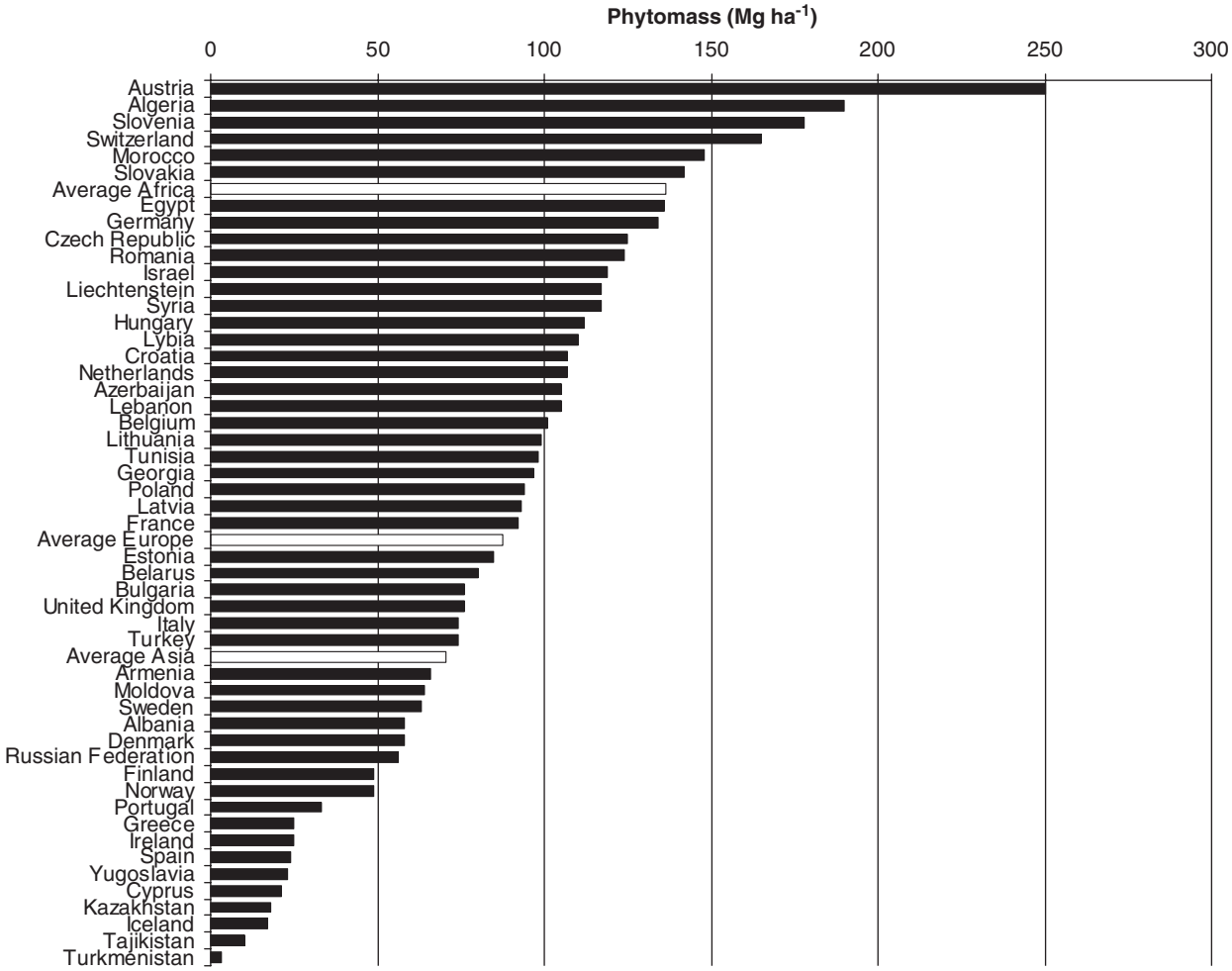

Fig. 2. Phytomass distribution within European and Asian countries for the year 2000.

Table 2
Mercury emission (kg yr^{-1}) from 1990 to 2004 estimated from standing phytomass in European, Asian and African countries

Country	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004
Albania	2.7	1.6	6.6	3.4	4.6	1.0	2.7	12.0	4.4	30.9	80.2	—	—	—	2.7
Armenia	1.1	0.7	0.7	0.7	0.7	1.1	0.7	0.1	2.4	0.4	0.3	1.0	—	—	1.1
Austria	5.6	1.5	3.7	3.1	1.6	0.9	0.8	0.6	2.6	0.2	1.2	—	—	—	5.6
Azerbaijan	0.7	0.7	0.3	0.3	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	—	—	0.7
Belarus	61.9	2.9	213.4	11.2	18.9	33.9	80.2	8.6	4.9	56.1	17.3	26.1	—	—	61.9
Belgium	0.2	0.6	0.2	1.2	0.6	0.7	16.4	3.2	0.3	0.0	0.0	0.0	—	—	0.2
Bulgaria	8.9	4.3	44.6	154.6	154.1	4.7	7.7	5.1	59.3	70.6	488.6	171.5	55.4	42.6	8.9
Croatia	9.9	42.0	107.6	186.8	50.8	24.1	73.7	81.3	384.1	72.5	816.9	424.5	—	—	9.9
Cyprus	3.4	0.3	0.0	0.2	0.4	0.2	0.3	0.4	1.3	0.0	18.9	11.4	5.2	5.5	3.4
Czech Republic	10.9	1.1	17.9	16.1	11.3	5.6	28.6	48.7	15.8	4.7	5.3	8.6	—	—	10.9
Denmark	0.9	0.9	1.8	0.1	0.0	0.0	0.4	0.1	0.2	0.0	0.0	0.0	—	—	0.9
Estonia	8.7	0.6	17.0	6.2	4.3	1.5	5.0	10.6	0.5	10.5	6.5	5.8	—	—	8.7
Finland	2.4	1.2	5.9	3.2	8.6	3.5	4.5	8.1	1.1	5.8	5.0	0.9	3.1	7.5	2.4
France	748.3	104.4	171.1	172.0	257.6	186.9	117.4	222.4	198.7	163.5	210.8	210.9	214.8	633.8	748.3
Georgia	1.4	1.1	1.1	1.1	1.1	0.0	2.2	1.1	1.1	1.1	1.1	1.5	—	—	1.4
Germany	14.2	13.8	73.7	22.4	16.7	8.9	20.7	9.0	6.0	6.2	8.7	1.8	1.8	19.7	14.2
Greece	108.1	36.5	199.9	151.3	162.1	76.2	70.9	143.3	315.8	53.3	467.6	51.4	112.7	9.5	108.1
Hungary	16.9	16.9	16.9	16.9	16.9	16.9	16.9	16.9	16.9	0.0	0.0	16.9	—	—	16.9
Ireland	2.4	1.9	1.6	1.5	0.8	1.3	1.3	1.3	1.3	1.3	1.3	1.3	—	—	
Israel	76.9	46.4	89.3	95.5	50.4	110.6	86.5	82.5	82.5	82.5	82.5	82.5	—	—	
Italy	1574.3	819.2	860.7	1617.6	1129.9	405.2	480.6	921.9	1289.2	747.0	1163.5	633.4	167.6	760.9	138.8
Kazakhstan	132.7	32.1	32.1	32.1	11.3	57.8	25.4	700.1	40.7	53.4	55.4	49.9	—	—	
Latvia	24.6	32.2	87.1	5.3	3.6	5.6	9.7	6.3	2.2	16.1	14.0	10.7	—	—	
Lebanon	—	—	—	—	—	—	5.5	5.1	71.6	17.9	—	—	—	—	
Libya	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Lithuania	4.6	0.6	9.6	3.4	3.3	3.6	4.3	1.5	1.0	2.4	2.4	1.2	—	—	
Moldova	0.4	0.1	0.2	0.0	1.5	0.0	0.0	0.0	0.1	0.1	0.1	0.4	—	—	
Morocco	35.1	65.7	42.7	51.0	100.6	116.3	19.6	63.7	30.7	28.0	—	—	—	—	
Netherlands	2.7	5.0	2.1	1.2	3.3	2.8	1.9	2.7	2.5	2.5	2.5	2.5	—	—	
Norway	0.5	2.9	7.5	1.2	1.3	0.6	2.8	4.8	1.6	4.7	9.4	5.3	—	—	
Poland	52.9	22.2	350.9	38.7	26.3	18.3	55.9	22.9	614.7	432.7	516.1	521.2	—	—	
Portugal	479.9	674.5	218.3	184.7	285.8	626.9	328.5	112.9	585.3	261.0	589.9	413.3	459.8	1573.4	478.8
Romania	6.3	3.8	6.5	5.2	3.3	2.0	2.9	0.6	10.1	5.3	50.1	21.8	—	—	
Russian Federation	9890.1	7063.7	7167.5	7529.1	4535.2	2903.1	14,500.4	6169.9	33,490.8	6028.6	11,940.0	7710.5	—	—	
Slovakia	3.1	3.7	3.7	8.2	1.5	1.4	3.5	0.6	0.5	8.9	14.4	4.9	9.5	24.9	
Slovenia	0.7	10.5	7.8	34.7	17.2	3.9	4.9	9.4	25.0	8.6	5.3	13.0	—	—	
Spain	548.5	657.8	281.1	240.1	1176.4	385.7	160.8	264.8	359.2	221.0	502.7	248.3	288.9	401.1	123.8
Sweden	—	—	41.0	7.1	17.5	2.8	15.4	45.1	21.1	21.1	21.1	8.8	—	—	
Switzerland	20.4	2.7	1.0	0.8	5.4	8.1	4.3	35.7	4.6	0.4	1.2	2.1	—	—	
Tajikistan	—	—	—	—	—	—	—	—	—	—	160.6	291.6	—	—	
Tunisia	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	15.6	
Turkey	107.7	62.7	101.9	107.1	173.7	38.8	120.1	49.8	56.1	48.1	218.4	61.3	—	—	
Turkmenistan	0.7	0.4	0.4	0.0	0.8	0.5	0.4	0.5	0.5	0.5	0.5	0.5	—	—	
United Kingdom	3.9	1.0	1.7	1.3	8.8	4.6	5.0	2.8	0.5	1.5	2.3	1.9	—	—	
Yugoslavia	—	4.0	5.1	17.8	4.1	4.3	10.1	—	8.5	4.6	20.5	8.9	—	—	
Total	13,990.4	9760.3	10,218.5	10,750.6	8289.0	5086.5	16,315.2	9092.6	37,732.5	8490.6	17,519.1	11,044.1	1334.8	3494.9	741.4

(—): data not available (revised from Pirrone et al., 2005).

preliminary assessment of Hg emissions for EU-25, Eur-Asia and African countries during the 1990–2004 period is reported in Table 2. Italy and Portugal show the highest annual average emissions with 821 and 458 kg yr^{-1} respectively, followed by Spain (394 kg yr^{-1}), France (247 kg yr^{-1}), Tajikistan (226 kg yr^{-1}), Poland (223 kg yr^{-1}) and Croatia (190 kg yr^{-1}). Mercury emissions in the remaining countries are in the range of 0.2 kg yr^{-1} (Moldova) to 140 kg yr^{-1} (Greece). The highest average annual contribution comes from the Russian Federation with 9910 kg yr^{-1} (with an annual peak of 33,490.8 kg in the year 1998!). As expected, fluctuations depend from the inter-annual burnt area with

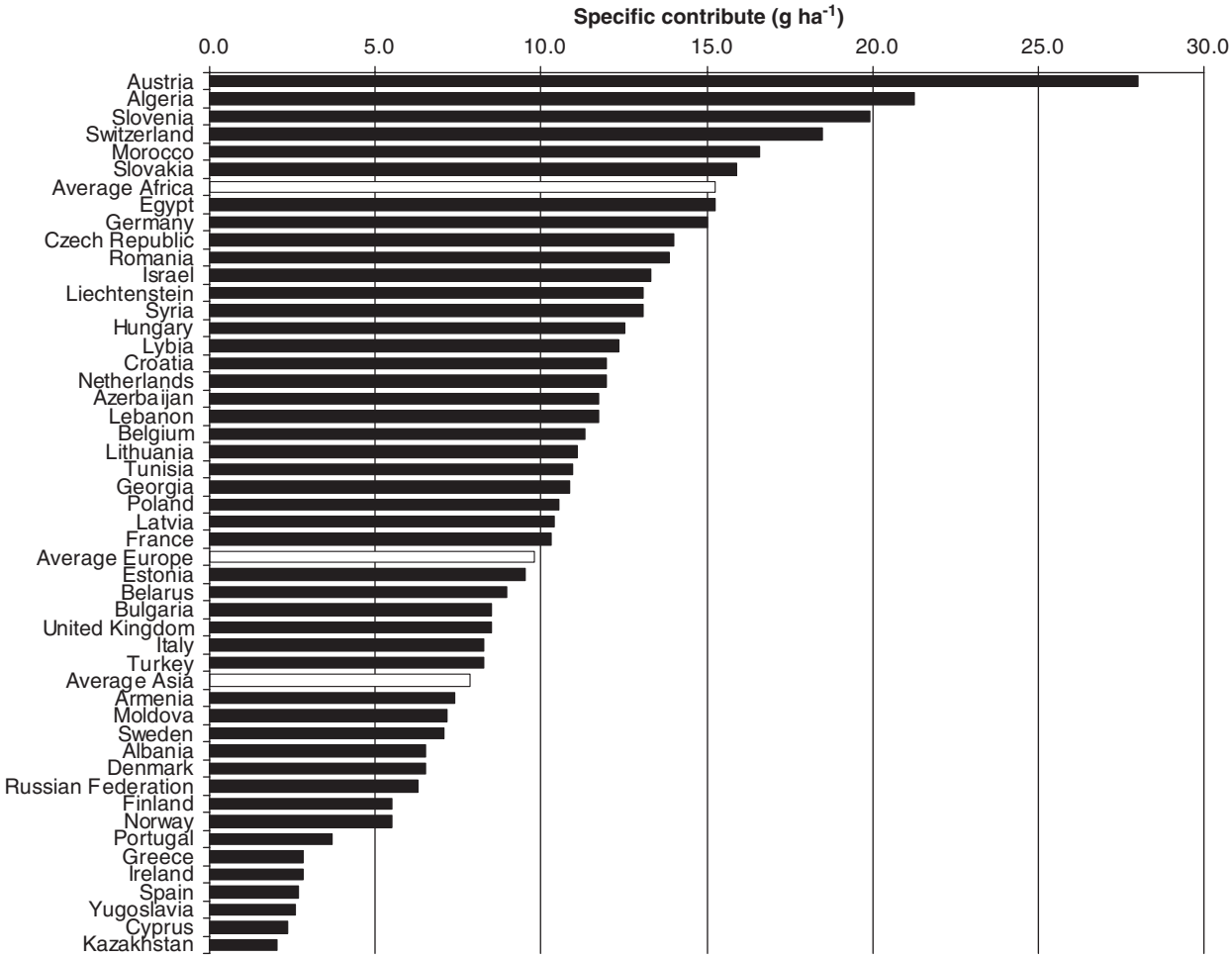

Fig. 3. Potential mercury emission rate assuming $112 \pm 17\,\mu g$ of mercury released per kg of dry phytomass.

a number of peak years which do not fall in the same year for different countries primarily due to changes in climatic conditions and socio-economic factors.

Assuming $112 \pm 17\,\mu g$ of mercury released per kg of dry phytomass, the mercury potential emission rate was estimated for European, Asian and North Africa countries (Fig. 3). Austria ($28.0\,g\,ha^{-1}$) shows the highest potential emission rate followed by Algeria ($21.3\,g\,ha^{-1}$), Slovenia ($20.0\,g\,ha^{-1}$) and Switzerland ($18.5\,g\,ha^{-1}$). The potential emission rates indicate the likely emission rate associated to the potential fire event.

On regional scale, Hg emissions in EU-25 ranges from 0.9 to $3.6\,Mg\,yr^{-1}$ ($2.4\,Mg\,yr^{-1}$ on average) (Fig. 4). Due to the highest contribution of Southern European countries (mainly Italy, France, Spain, and Portugal) similar values are estimated for the Mediterranean region ($2.0\,Mg\,yr^{-1}$ EU countries, $0.2\,Mg\,yr^{-1}$ Eastern European countries, $0.1\,Mg\,yr^{-1}$ Asian countries and $0.1\,Mg\,yr^{-1}$ African countries). These estimates suggest that the contribution from forest fires represents a small fraction of the total atmospheric mercury emissions in the region (years it ranges between 0.57 and 1.51%) (Table 3), though its trend is upward compared to the declining trend in anthropogenic emissions (Pirrone et al. 2001; Pacyna et al., 2003, 2005; Pirrone et al., 2005) and to the large variability in burnt areas. The variation of the inter-annual burnt area between 4000 and $8000\,km^2$ (Schelhaas and Schuck, 2002) suggests that the amount of burnt surface is substantially stable and the contribution from forest fires to the global mercury budget can be important in the near future.

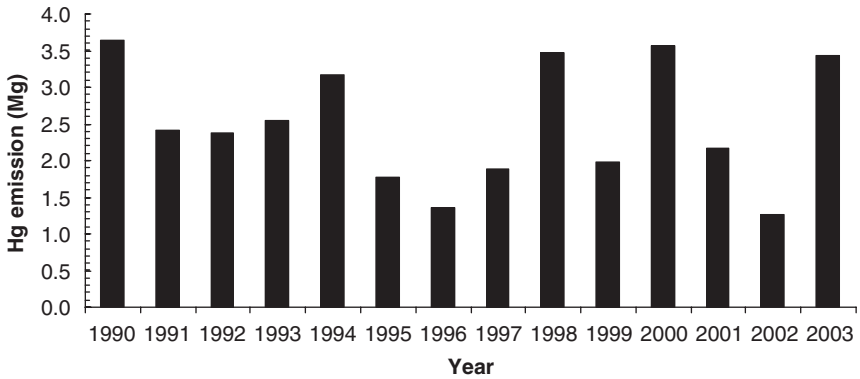

Fig. 4. Annual mercury emissions from forest fires in EU-25 countries, Norway and Switzerland from 1990 to 2003.

Table 3
Comparison between Hg emissions ($Mg\,yr^{-1}$) from anthropogenic sources and forest fires in EU-15 for the 1980–2000 period

	1980	1985	1990	1995	2000
Anthropogenic sources	860	726	627	338	239
Forest fires	0	0	3.6	1.8	3.6

Table 4 reports Hg emissions for different ecosystems in the world. Sigler et al. (2003) estimated 3.5 $Mg\,yr^{-1}$ for Canadian boreal forests considering the Hg release per unit of fuel, whereas Friedli et al. (2003) estimated 60 $Mg\,yr^{-1}$ for temperate/boreal ecosystems. For the primary forest in the Amazonian region Veiga et al. (1994), de Lacerda (1995) and Roulet et al. (1999) estimated a total mercury release between 6 and 88 $Mg\,yr^{-1}$. On global scale Brunke et al. (2001) derived Hg emissions from CO and CO_2 emission data which was between 380 and 1330 $Mg\,yr^{-1}$ (average 590 $Mg\,yr^{-1}$), whereas those estimated by Friedli et al. (2003) were about 900 $Mg\,yr^{-1}$. These differences indicate that Hg released from forest fires may be highly dependent on forest type and their proximity to anthropogenic sources. It is important to point out that our evaluation of mercury emissions from this particular source does not take into account the contribution from agricultural practice, which uses fire for clearing the land or for improving the pasture for livestock grazing, therefore, the evaluation of Hg emissions reported here may be underestimated.

3.2. Mercury emissions in the Russian Federation

Mercury emissions from the Russian Federation were estimated for the 1996–2002 period using the RS data. The southernmost part of the federation (Krasnoyarsk, Irkutsk, Amur and Chita districts region) show the hot-spot areas (Fig. 5). Annual trend is comparable with that estimated by ground-based data showing a peak in 2006 with ~75,000 km^2 of burnt area (Table 5). The phytomass consistency (Fig. 6) is distributed following the climatic belts and socio-economic pressure that differ between the Western and the Eastern part of Russia. The former has experienced a significant increase in the average growing stock while the latter experienced an important decrease due to an intensive exploitation of forests. The estimate of phytomass (average 66 $Mg\,ha^{-1}$) and burnt area allowed to identify several hot-spots with an average of Hg emission rate of ~8 g of mercury per hectare.

During the period 1996–2002 Hg emissions were in the range of 3.4–24.8 $Mg\,yr^{-1}$ with an average value of 16 $Mg\,yr^{-1}$ which is higher (>20%) than that derived from ground-based data (13.3 $Mg\,yr^{-1}$) (Table 5). However, the uncertainty associated with the assessment of burnt areas and phytomass amount may be significant in the overall regional mercury emissions estimate. Indeed, burnt areas derived from RS data were nearly double (236,409 km^2) compared to that derived from ground-based data (127,296 km^2), whereas the difference in phytomass amount estimated with ground-based (56 $Mg\,ha^{-1}$) and RS (66 $Mg\,ha^{-1}$) data is below 20%. As consequence, differences in mercury emission estimates from RS and ground-based data are primarily related to differences in phytomass density and burnt area. The discrepancy between burnt area estimated with different methodologies (i.e. ground or aerial versus satellite observations) is well known (GFMC, 2003). Also sensor typology affects the phytomass estimate

Table 4
Mercury emission from forest fires estimated in this work compared with that reported in literature

Forest	Total Hg release ($Mg\,yr^{-1}$)	Average area burnt ($10^6\,ha\,yr^{-1}$)	Average fuel consumption ($Tg\,yr^{-1}$)	Hg release per unit of fuel burnt ($ng\,g^{-1}$)	Reference
Temperate/boreal					
Canadian boreal	3.5	2.3	55	63.41	Sigler et al. (2003)
Russian boreal	13.3	2.1	119	111.95	This work[a]
Russian boreal	16.1 (3.4–24.8)	3.9	260	61.92	This work[b]
Temperate/Mediterranean (Europe)	2.4 (0.9–3.6)	0.5	46	52.75	This work[a]
Mediterranean (Europe/Africa)	2.3 (0.4–4.0)	0.5	45	51.11	This work[a]
Temperate/boreal	59.5	—	530	112.05	Friedli et al. (2003)
Boreal forests	22.5	5.0–15.0	240	93.75	Sigler et al. (2003)
Tropical					
Tropical (Amazonian)	88.0	3.0–5.0	1404	62.68	Veiga et al. (1994)
Tropical (Amazonian)	17.0	1.5–2.1	486	34.98	de Lacerda (1995)
Tropical (Amazonian)	6.0–9.0	2.0–3.0	843	8.90	Roulet et al. (1999)
World					
All forests	20.0	—	—	—	Nriagu (1989)
All forests	930 (510–1140)	—	621	1497.58	Brunke et al. (2001)[c]
All forests	590 (380–1330)	—	3460	170.52	Brunke et al. (2001)[d]
Global burning biomass	853.0	—	7432	114.77	Friedli et al. (2003)

Estimate based on:
[a]Ground-based data (1996–2002).
[b]Remote sensing data (1996–2002).
[c]Hg/CO emission ratio.
[d]Hg/CO_2 emission ratio.

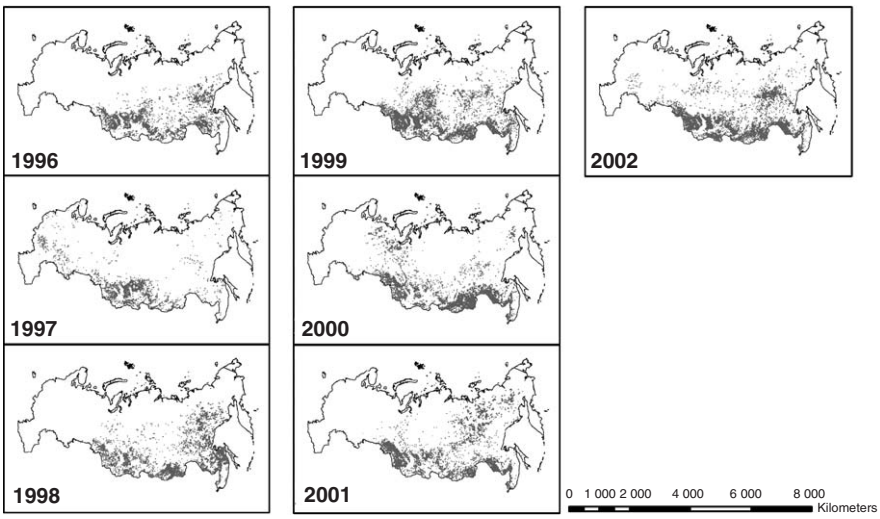

Fig. 5. Yearly trends of burnt areas in Russia from 1996 to 2002.

(Lee et al., 2004) and quality data made available to the public can be a significant source of uncertainty (Karpachevskiy, 2004) (Fig. 7).

Russian forests are mostly boreal forest type (Conard and Ivanova, 1997) enabling the comparison with Sigler et al. (2003) and Friedli et al. (2003) estimates (Table 4). Our estimate ($16.1\,Mg\,yr^{-1}$) is lower than that reported by Sigler et al. (2003) ($22.5\,Mg\,yr^{-1}$) and by Friedli et al. (2003) ($59.5\,Mg\,yr^{-1}$) by a factor of 1.4 and 3.7, respectively.

Table 5
Mercury emission ($Mg\,yr^{-1}$) and burnt areas (km^2) in the Russian Federation estimated from ground-based and remote sensing data for the 1996–2002 period

Year	Ground-based estimates		Remote sensing estimates	
	Burned area (km^2)	Hg emission ($Mg\,yr^{-1}$)	Burned area (km^2)	Hg emission ($Mg\,yr^{-1}$)
1996	23,119	14.5	35,768	13.4
1997	9837	6.2	7588	3.4
1998	53,397	33.5	60,457	22.8
1999	9612	6.0	36,635	14.6
2000	19,037	11.9	55,693	20.2
2001	12,294	7.7	40,268	13.3
2002			75,229	24.8
Averages				
Burnt phytomass ($Mg\,ha^{-1}$)		56		66
E_f ($mg\,Mg^{-1}$)		112		112
Hg emissions ($g\,ha^{-1}$)		6.3		7.4
Hg release ($Mg\,y^{-1}$)		13.3		16.1
Burnt area (km^2)		21,216		39,402

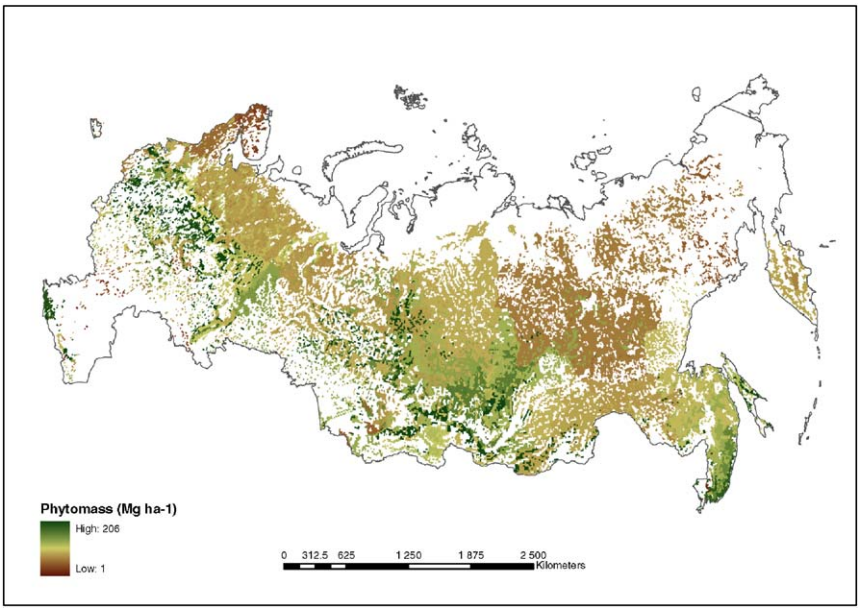

Fig. 6. Spatial distribution of phytomass in Russian forests in 2000.

An additional comparison based on the mercury–carbon ratio was performed. The Hg–C ratio has often been used to estimate Hg emissions from wildfires (Brunke et al., 2001; Friedli et al., 2003; Sigler et al., 2003). In detail, the Hg/CO ratio during a forest fire was estimated as reported in Table 6a and the amount of CO released to the atmosphere can be estimated considering the plume speciation data reported in Table 6b (Ward et al., 1992; Ward, 1995; Ward et al., 1996; Battye and Battye, 2002).

Carbon emission estimates for forest fires in Russia were referred to 2002, considered as reference year. Carbon released to the atmosphere from forest fires was 210 Tg (1 Tg = 10^9 g), which implies a release of 49.1 Tg of CO. The estimate of mercury emissions is obtained by multiplying the emission ratios with the CO amount (Table 7). Our estimates (24.8 $Mg\,yr^{-1}$) are similar to that proposed by Sigler et al. (2003) (30.3 $Mg\,yr^{-1}$) whereas higher differences have been found by comparing our estimate

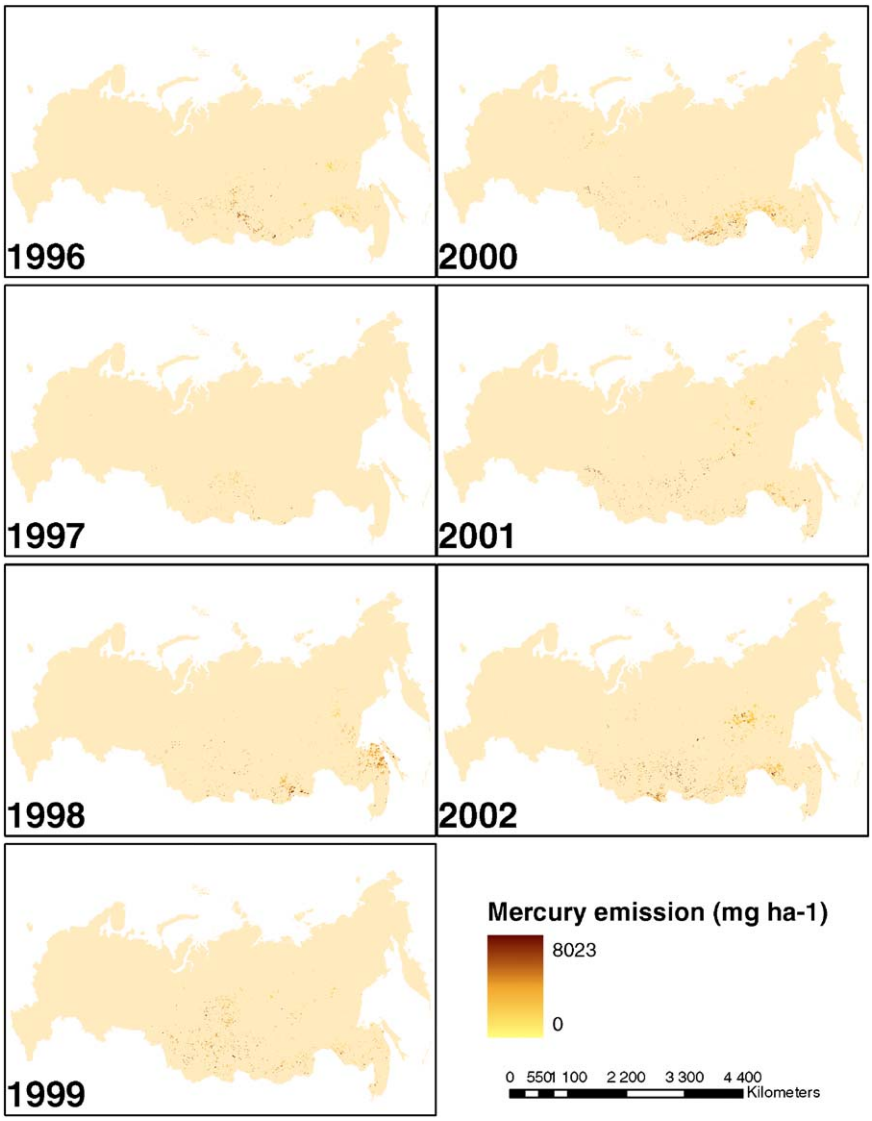

Fig. 7. Annual Hg emissions from forest fires from 1996 to 2002.

with that of Brunke et al. (2001) (73.9 $Mg\,yr^{-1}$) and Friedli et al. (2003) (105.5 $Mg\,yr^{-1}$).

3.3. Uncertainty

The uncertainty associated to our estimates is related to several factors which include emission factor variability with forest type and time of the year, the evaluation of burnt area, the estimate of burnt phytomass (combustion efficiency), the forest fire dynamics and quality of data coverage.

The Emission Factor used in our assessment was derived from in-plume measurements of forest fires over a conifer forest (Black spruce), which shows a 15% variation ($112 \pm 17\,\mu g\,kg^{-1}$) (Friedli et al., 2003). From the above, the average Hg emission in EU-25 (2.4 $Mg\,yr^{-1}$) ranges from 2.1 to 2.8 $Mg\,yr^{-1}$ (considering the ground-based data) and 13.7–18.5 $Mg\,yr^{-1}$ (considering the 16 $Mg\,yr^{-1}$ as average RS estimates for the Russian Federation). Because the mercury content in Mediterranean vegetation is driven by the high deposition rate (5–10 $\mu g\,m^{-2}$) (Ryaboshapko et al., 2005) we may expect a higher E_f than that assumed in our assessment.

The errors related to the input data depend from the data quality of burnt areas. In particular, unburnt areas within a fire area are often

Table 6
Mercury-carbon emission ratios (a) and carbon content in forest fire plumes (b) found in literature

		Reference
(a) Hg–CO *emission ratios* ($ng\,g^{-1}$)		
1500 ± 150		Brunke et al. (2001)
2150		Estimated from Friedli et al. (2003)
617		Sigler et al. (2003)
(b) *Carbon content*		
Compound	Ratio	
CO_2	0.888	
CO	0.100	Ward et al. (1996); Battye and Battye (2002)
CH_4	0.012	

Table 7
Mercury emission estimates (minimum, average and maximum) based on 49.1 Tg of CO released by forest fires in the Russian Federation and on emission ratios reported in Table 6

Reference	Hg/CO emission ratios ($ng\,g^{-1}$)	Hg released ($Mg\,yr^{-1}$)		
		Min	Ave	Max
Brunke et al. (2001)	1500 ± 150	63.8	73.9	84.0
Estimated from Friedli et al. (2003)	2150	89.7	105.5	121.4
Sigler et al. (2003)	617	27.3	30.3	33.3
Estimated in this work	855		24.8	

The 2002 was assumed as reference year.

unmapped, leading to an overestimate of the total burnt area. Also the definition of forest area leads to a misinterpretation of the information, and discrepancies were found when ground-based data provided from different data set were compared (i.e. FAO, National Statistic Offices). In the case of RS data, accuracy of fire mapping depends from scene cleanness, spatial resolution, vegetation density, fire size, fire severity and vegetation moisture content (Remmel and Perera, 2001). The error associated with this assessment was found within 15–30%. The combination of RS and GIS represents a powerful tool to improve Hg emission estimates from forest fires. This technique is more accurate than that based on ground-based data because the phytomass amount and burnt areas are estimated at pixel level. The resolution depends on the pixel dimension that is a characteristic parameter of the satellite sensor and is related to several factors (i.e. area dimension, scene cost) (Cinnirella, 2003). Advanced Very High Resolution Radiometer (AVHRR) is used to generate global daily data set on phytomass burning activity (Hugh and Lambin, 1998; Barbosa et al., 1999; Levine et al., 2004), while Landsat Advanced Thematic Mapper (ETM+) offers forest cover type classification and stand phytomass with 15×15 m resolution (Ecoleap, 2004). Moreover, the Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Terra satellite is a well suitable instrument when used in a multi-temporal mode. Finally, the combination of remotely sensed images (i.e. MODIS, AVHRR, ETM+) may improve the estimate of both phytomass and burnt areas (Csiszar et al., 2004).

The stock of phytomass estimate is affected by several errors; some of them are originated by the estimate of basic phytomass content of forests (Lakida et al., 1997; Alexeyev and Birdsey, 1998). Growing stock of forests is underestimated by 10%, however the timber density may affects the final estimate because of its depletion process and carbon losses. Alexeyev and Birdsey (1998) estimate the timber density with an accuracy of $\pm 30\%$. As consequence the Hg emission estimate with RS data for the Russian federation is revised considering a $\pm 8\,Mg\,yr^{-1}$ uncertainty, which leads to an increase/decrease of the emission by 50%. It is well recognized that the combustion of phytomass depends upon several factors including fuel type, fuel load, fuel chemistry, fuel moisture content, packing ratio for the fuel particles, surface area to volume ratio and method of ignition. Fire dynamics and combustion efficiency must be considered in the overall uncertainty estimate. Differences on fire typology (surface, crown, and total) of forested areas change the amount of burnt phytomass leading to a lower or higher Hg emission. The latter was combined with the combustion efficiency for which is generally assumed 100% consumption for litter and herbaceous phytomass, 60% consumption for shrub and tree regeneration, 100% consumption for canopy foliage and 50–65% consumption for canopy fine branchwood. From the above, arise the consideration that dramatic changes in Hg emission derive from changes in the combustion efficiency and that a large impact on the E_f should be expected

in the Mediterranean environment where 90% of fuel is consumed during a forest fire.

The forecast of burnt areas is a key step in the interpretation and prediction of seasonal and inter-annual emission variations in forest fire occurrence. Therefore, the correlation of long-term atmospheric anomalies and forest fire activity may allow mercury emissions prediction from this particular source. Results from this study are certainly limited because of the generalization of the E_f over a wide region but the main source of error comes from the burnt area estimate followed by the burnt phytomass estimate. Information on E_f given by Friedli et al. (2003) show that a significant amount of mercury is originated from soil, however much has to be done to improve our capability to better estimate the Emission Factor for each type of forest, season and region.

4. Conclusions

The aim of this paper was to present a first assessment of mercury emissions from forest fires in the Mediterranean Region and in the Russian Federation. The approaches were based on ground surveys of burnt areas that lead to a country based estimate and on RS survey of burnt areas that provides a cell-by-cell estimate. Annual mercury emissions have been estimated in the range of 0.9–3.6 $\mathrm{Mg\,yr^{-1}}$ for Europe and 0.4–24.8 $\mathrm{Mg\,yr^{-1}}$ for the Russian Federation, respectively.

The uncertainty was discussed in relation with the assumption of an undifferentiated emission factor for each forest type and time of the year, the evaluation of burnt area, the estimate of burnt phytomass (combustion efficiency), the forest fire dynamics and quality of data coverage. The uncertainty lead to the conclusion that mercury emissions from wildfires in the Mediterranean and in the Russian Federation represent a minor contribution compared to anthropogenic emissions and estimates should be interpreted as a first attempt to estimate Hg emissions from this particular source.

On the other hand estimates based on RS approach give a better spatial and temporal resolution of burnt areas and biomass amount. In addition, the cost of satellite images imposes the limit to have updated data on Hg emission at low cost. Nevertheless, the usefulness of the methodology is clear and quantitative estimates of mercury emissions from forest fires might be extended to all countries when the satellite data will became accessible. Indeed, estimates with RS and GIS techniques are fundamental for modellers that use gridded data for their assessments.

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