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# Ideas and Perspectives: On the emission of amines from terrestrial vegetation in the context of atmospheric new particle formation

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## Abstract

In this article we summarise recent science, which shows how airborne amines, specifically methylamines (MAs), play a key role in atmospheric new particle formation (NPF) by stabilising small molecule clusters. Agricultural emissions are assumed to constitute the most important MA source, but given the short atmospheric residence time of MAs, they can hardly have a direct impact on NFP events observed in remote regions. This leads us to the presentation of existing knowledge focussing on natural vegetation-related MA sources. High MA contents as well as emissions by plants have already been described in the 19th century. Strong MA emissions predominantly occur during flowering as part of a pollination strategy. The behaviour is species specific, but examples of such species are common and widespread. In addition, vegetative plant tissue exhibiting high amounts of MAs might potentially lead to significant emissions, and the decomposition of organic material could constitute another source for airborne MAs. These mechanisms would provide sources, which could be crucial for the amine's role in NPF, especially in remote regions. Knowledge about vegetation-related amine emissions is, however, very limited and thus it is also an open question how Global Change and the intensified cycling of reactive nitrogen over the last 200 years have altered amine emissions from vegetation with a corresponding effect on NPF.

## 1 Introduction

Amines comprise a vast range of nitrogenous organic compounds with aliphatic methylamines (MA; monomethylamine = MMA, dimethylamine = DMA, trimethylamine = TMA) representing the most common (Ge et al., 2011a) and comparably most studied airborne species. Amines have widely been identified in aerosol particulate matter and wet deposition (Neff et al., 2002; Ge et al., 2011a; Healy et al., 2014; Wang et al., 2015).

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New aerosol particle formation (NPF) from gas-phase precursors occurs frequently in the atmosphere, approximately constituting 50 to 80 % of the global aerosol load and 30 to 50 % of cloud condensation nuclei (Merikanto et al., 2009). Estimates of the global secondary aerosol budget are highly uncertain due to uncertainties in gas-phase precursor contributions to NPF and limited mechanistic comprehension (Spracklen et al., 2011). While NPF rates observed in the field do not comply with the classical process-understanding involving sulphuric acid (SA) and water (Kirkby et al., 2011; Chen et al., 2012), it has recently become increasingly clear that the ternary system of SA and water, together with neutralising compounds like ammonia (NH<sub>3</sub>) or amines, is a key system in NPF (Chen et al., 2012; Almeida et al., 2013; Kurten et al., 2014). Amines can also form stable molecular clusters with methanesulfonic acid (Dawson et al., 2012). In addition, the oxidation of organic compounds contributes to NPF (Riccobono et al., 2014).

Despite their generally low atmospheric concentrations (Ge et al., 2011a), Almeida et al. (2013) have shown that amines take an important role in NPF. Their main finding is that extremely small MA mixing ratios (> 3 ppt DMA in their case) suffice to efficiently enhance NPF rates, and that atmospheric observations of NPF rates are only consistently explained taking amines into consideration. This is consistent with the current mechanistic understanding. The NPF enhancement by addition of MAs is most significant in situations with clean air, essentially when only little NH<sub>3</sub> is present to contribute to SA neutralisation.

The indirect effect of anthropogenic aerosols represents the largest uncertainty on climate radiative forcing according to IPCC (2013). Almeida et al. (2013) note how the recent findings “show that the uncertainty is even greater than previously thought, because extremely low amine emissions – which have substantial anthropogenic sources and have not hitherto been considered by the IPCC – have a large influence on the nucleation of sulphuric acid particles. [...] If amine emissions were to spread into pristine regions of the boundary layer where they could switch on nucleation, substantial increases in regional and global cloud condensation nuclei could occur”.





ing its (SA-dependent) threshold concentration, the NPF rate solely depends on the availability of SA (Kurten et al., 2014).

In the atmosphere, SA is continuously formed in-situ via oxidation of sulphur dioxide (Boy et al., 2005). Typical SA mixing ratios cover a range from 0.005 to 5 ppt (Eisele and Tanner, 1993; Petäjä et al., 2009; Almeida et al., 2013). Hence, the emissions of the alkaline gases determine, to a large part, the potential for NPF. In polluted air masses, for example close to intensive agricultural activities or at urban environments, usually enough airborne  $\text{NH}_3$  and MAs exist (Ge et al., 2011a; Sintermann et al., 2014) to favour high NPF rates. The high concentrations of existing particles often hinder NPF by acting as coagulation sink for newly formed clusters (Kerminen et al., 2001; Dal Maso et al., 2007; Westervelt et al., 2014). In clean pristine air, however, the addition of small amounts of an atmospheric base is expected to promote NPF. This much more efficiently, if that base is a MA rather than  $\text{NH}_3$ . Thus, MA emissions in regions with clean air will have a strong impact on NPF – especially when they are not coupled to larger  $\text{NH}_3$  emissions.

In the environment, the availability of emitted MAs for NPF is constrained by their removal via dry and wet deposition, gas-phase oxidation – mainly by the hydroxyl radical – (Ge et al., 2011a; Lee and Wexler, 2013), and condensation onto pre-existing particles. The latter is an efficient removal process yielding a range of MA lifetimes depending on the aerosol number-size distribution and the assumed uptake coefficient (defined as the fraction of molecules being permanently removed from the gas phase by collision with an existing particle). Sintermann et al. (2014) roughly estimate a condensation sink lifetime of TMA of 0.5 and 17 min for a high and lower aerosol burden, respectively, based on an uptake coefficient of 1. Using a chemistry-transport model, approximating amine emissions by scaling with those of  $\text{NH}_3$  (the approach taken by Schade and Crutzen, 1995), including oxidation, deposition, and the condensation sink, Yu and Luo (2014) derive atmospheric MA lifetimes. With a maximal uptake coefficient of 0.03, they calculate atmospheric residence times of approximately 1 h over Europe, eastern Asia, and the eastern US compared to 3 to > 5 h in more re-

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remote regions. According modelled MMA/DMA/TMA concentrations span approximately 5/2/10 ppt over India, 2/0.5/5 to 5/1/10 ppt over China and parts of Africa and South America, 0.2/0.1/0.5 to 2/0.5/5 ppt over the major parts of the other continents to 0.05/0.02/0.1 to 0.5/0.2/0.5 ppt in more remote regions (excluding the polar areas).

5 Comparing their results to existing concentration measurements they suggest, however, that emissions further away from urban areas, and hence concentrations, might be substantially underestimated. This would be emphasized even more strongly by the finding that the estimated agricultural amine emissions do not scale with a constant ratio throughout all  $\text{NH}_3$  emission stages (Kuhn et al., 2011; Sintermann et al., 2014).  
10 In conclusion, the short atmospheric residence time of MAs implies that only local to regional emissions are crucial in promoting NPF. Emissions into clean air would spread the MA potential for NPF further than in situations with higher aerosol burdens.

Measurements of atmospheric MAs are challenging and reports of concentrations in regions away from potential urban, agricultural, or marine sources are rare. Simultaneous observations of MAs and  $\text{NH}_3$  and NPF in remote areas would be essential to characterise the corresponding interactions. As far as we know, the only campaigns at comparably remote continental regions have been carried out at the Finnish boreal forest site Hyytiälä (Hari and Kulmala, 2005). Sellegri et al. (2005) found 34 to 80 ppt TMA (and 12 ppt DMA which was below the detection limit) during a short period in spring 2003, whereas Kieloaho et al. (2013) determined ambient DMA + ethylamine (EA) and TMA + propylamine (PA) from May to October 2011. Highest concentrations DMA + EA and TMA + PA amounted to  $157 \pm 20$  and  $102 \pm 60$  ppt, respectively, in September/October, which were thought to originate from litterfall-emissions. DMA + EA exhibited somewhat smaller concentration peaks ( $> 50$  ppt) at the beginning of June and in July/August. Diethylamine (DEA) from unassigned sources was also measured and peaked in the summer months ( $15.5 \pm 0.5$  ppt). The observation of NPF events was only moderately correlated to the occurrence of the measured amines. As shown by Hellén et al. (2014), the amines were generally more abundant at the boreal forest site than in the urban environment of Helsinki during the

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5 same period. It appears striking that the prevailing amine concentrations at the bo-  
real forest exceed the threshold values given by Almeida et al. (2013) and Jen et al.  
(2014) at which the maximum NPF rate could occur. Schade and Crutzen (1995) es-  
timated an average MA : NH<sub>3</sub> emission ratio of 0.7 % from agricultural sources, which  
10 are assumed to dominate the anthropogenic MA source and likewise, Sintermann et al.  
(2014) measured a maximum TMA emission ratio of 1 %. By contrast, simultaneous  
measurements of MAs and NH<sub>3</sub> at a coastal-rural site, with potential marine source  
influence, indicated time averaged concentration ratios DMA : NH<sub>3</sub> = 1.6 to 3.5 % and  
TMA : NH<sub>3</sub> = 0.75 to 3.9 % (Freshour et al., 2014). In another study at two rural forested  
15 areas, the MA : NH<sub>3</sub> ratio did not exceed 1 %, but correlation with isoprene led to spec-  
ulation about a vegetation source (You et al., 2014). Despite high measurement uncer-  
tainties (Freshour et al., 2014), such elevated MA concentrations and higher ratios to  
NH<sub>3</sub> in comparatively remote situation compared with agricultural environments, could  
point towards vegetation as another source for MAs.

### 15 3 Vegetation as a potentially significant source for gas-phase amines

It has been observed that a diverse group of several plant species exhibits exceptionally  
high amounts of MAs associated (a) either generally with their vegetative tissue (e.g.  
foliage), or (b) with blossoms during flowering (Smith, 1971, 1975). These species are  
MA reservoirs representing strong potential MA emitters. Wicke (1862) was among the  
20 first to describe the emission of TMA from plants. He observed the occurrence of TMA  
in condensed transpiration of *Chenopodium vulvaria*, known for its fish-like smell which  
is characteristic of TMA. Remarkably, he saw some kind of “fog” emerging when expos-  
ing a HCl-coated glass rod to the air close to the plant. He might indeed have been the  
first scientist to witness the effects of NPF from TMA neutralising the acid. At that time it  
25 had been known that also pears (*Pyrus communis*), hawthorn (*Crataegus monogyna*),  
and the rowan (*Sorbus aucuparia*) contain TMA in their blossoms (Wicke, 1862). Later,  
the extremely high TMA content of *Chenopodium vulvaria* was established by Cromwell

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(1950) and Cromwell and Richards (1966), who followed the works of Power and Chesnut (1925); Vickery (1925); Klein and Steiner (1928); Steiner and Löffler (1929); Vickery (1932), among others. By then, the range of higher plants with a high MA content in foliage or blossoms had been extended to a few more species. Further on, Stein von Kamienski (1957a, b, 1958) and Steiner (1966) provided measurements comprising the identification of MAs in foliage and especially blossoms of various plants (trees, bushes, herbs), a larger number of fungi, and even some lichen and moss, demonstrating that enrichment of volatile amines is widespread among some families (like Araceae, Caprifoliaceae, Cornaceae, Rosaceae, Umbelliferae) and almost not found in other families (Labiatae, Papilionaceae) (Smith, 1971). Volatile amine abundance is highly species-specific and may depend on phenology and site (Stein von Kamienski, 1957a; Steiner, 1966; Smith, 1971). Smith (1971) reviewed the state of knowledge about amines and plants at that time. MMA, DMA, and TMA have commonly been found in the amine-enriched vegetation. Table A1 (Appendix) summarises species associated with MAs found in the literature. Some examples of higher plant species, which are widespread in Europe, include: *Castanea sativa*, *Cornus sanguinea*, *Crataegos laevigata*, *Crataegus monogyna*, *Mercurialis annua*, *Mercurialis perennis*, *Pyrus communis*, *Sorbus aucuparia*, among others.

It is not entirely clear which pathways lead to MA creation within such plants (Steiner, 1966; Smith, 1971). In principle, formation pathways are the decarboxylation of amino acids and transamination from aldehydes (Cromwell and Richards, 1966; Steiner, 1966; Steiner et al., 1967; Smith, 1971, 1975; Wink and Hartmann, 1981; Dey et al., 1997; Farquhar et al., 1983; Bagni and Tassoni, 2001; Dudareva et al., 2013). Most measurements addressed the MA content within the tissue of vegetation parts such as fruiting bodies, foliage, and especially blossoms. However, the MAs are also released into the gas phase. Often the characteristic smell was observed, indicating that amines were also being emitted from the living plant. In fact, many plants use the chemical mimicry effect of aminoid fragrances, resembling excrements and decay, in order to attract dung and carrion insects, such as flies and beetles, for pollination. The scents in-

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volve a complex composition of many different volatiles with various nitrogen-containing compounds (Dobson, 2006; Abrol, 2012; Woodcock et al., 2014). It has been recognised that MA volatilisation from blossoms plays an important role in that pollination mechanism (Smith, 1971). It might also be that amines serve as antioxidants (Larson, 1988).

At least 15 different amines have been identified as floral scent compounds, and generally the quality and quantity of scents varies spatially and temporally, from plant to plant, and between species (Dudareva and Pichersky, 2006; Knudsen and Gershenzon, 2006). The blossoms are usually, but not necessarily, of white, green, or red-brown colour and often exhibit the characteristic amine odour (Stein von Kamienski, 1957a). One example of plants using MAs as part of a pollination strategy is the hawthorn (here: *Crataegus monogyna* and *Crataegus laevigata*). Hawthorn shows the characteristic scent of TMA (El-Sayed, 2014). It is striking that already in medieval times hawthorn branches were a symbol of the Black Death, since the hawthorn's fragrance mimics decay (Vickery, 1995).

Our own dynamic chamber measurements using Chemical Ionisation Mass Spectrometry (CIMS) highlight strong TMA emissions from freshly cut twigs with blossoms of *Crataegus laevigata* and *Cornus sanguinea* (Fig. 1). These measurements originally had an explorative intention, but we tentatively derive emission estimates. Highest TMA emissions from the approximately 45 *Crataegus laevigata* blossoms were in the range 0.019–0.038 nmols<sup>-1</sup> and peaked between 0.011 and 0.022 nmols<sup>-1</sup> for *Cornus sanguinea* (5 umbels). 1000 (as an arbitrary number) blossoms or 100 umbels, respectively, would thus have emitted up to 0.84 and 0.44 nmols<sup>-1</sup>. As a comparison, the TMA emissions from 1 square meter covered with a mix of farm animal excrements, as a very strong TMA source, have been determined to be of the order of 10 nmols<sup>-1</sup> (Sintermann et al., 2014). It is striking that the NH<sub>3</sub> volatilisation from the blossoms is much lower than that of TMA, implying that in the absence of another significant NH<sub>3</sub> source (like agriculture), vegetation-related MA emissions might be an important pathway providing atmospheric bases available for NPF.

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*pseudoacacia*, *Acer platanoides*, *Acer pseudoplatanus* yield high choline concentrations in phloem exudate (Kaiser and Ziegler, 1987). Betaines, which contain TMA, are widely distributed among higher plants with a couple of species accumulating betaines (Blunden et al., 2005). Polyamines with manifold physiological functions, such as putrescine, spermidine, and spermine, are highly abundant in plants (Moschou et al., 2012).

During the microbial decomposition of organic matter, volatile amines can be formed. Amino acid decarboxylation is featured in the soil (Yan et al., 1996) and, consequently, MAs and other amines are present in dissolved organic nitrogen derived from forest soils (Yu et al., 2002). The liberation of amines, including MMA, has been observed from bacterial cultures (Claeson, 2006). One example of a controlled and intensified way to degrade organic material is composting. In the headspace of composting facilities high concentrations of amines, especially TMA, have been found (Maris et al., 1999; Mao et al., 2006; Tsai et al., 2008). Emission of  $\text{NH}_3$  from senescent and decaying plant material has been described and corresponding  $\Gamma$  values have been evaluated (David et al., 2009; Mattsson et al., 2009; Massad et al., 2010a; Flechard et al., 2013). To which extent a similar mechanism also leads to MA emissions in the field needs to be investigated.

## 4 Context and conclusions

Contemplating very recent and quite long standing science, we have presented evidence that terrestrial vegetation, especially during flowering, could constitute an amine source that has the potential to unfold a notable effect on NPF. This kind of source most likely prevails during spring. Other vegetation-related amine emissions, such as those from non-flowering plants, fungi, or decomposing organic matter probably exhibit a wider amplitude during the year with a concentration towards autumn. In Scandinavian boreal forests, NPF is a frequent phenomenon occurring approximately during 25 to 50 % of all days. These characteristics exhibit two peaks: one more pronounced dur-

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ing March to May/June and another smaller maximum around September (Dal Maso et al., 2005, 2007; Kulmala et al., 2012). Biogenic VOC emissions and the corresponding oxidation of terpenoids are thought to influence this pattern (Kulmala et al., 2004). Advection of clean air exhibiting a small condensation and coagulation sink favours NPF events (Dal Maso et al., 2007), which underlines the importance of the contribution of biogenic emission in remote areas. It currently remains speculative how biogenic amine emissions play a quantitative role in this story, but amines have come to be considered a key element in the stabilisation of small clusters, which represents a bottleneck to NPF.

Through the invention of the Haber-Bosch process in 1913, humans have greatly intensified the reactive nitrogen ( $N_r$ ) cycle (Galloway et al., 2003) with an increase of the emissions from  $13 \text{ Tg Nyr}^{-1}$  in 1860 to  $46 \text{ Tg Nyr}^{-1}$  in 1995 and a projection to  $82 \text{ Tg Nyr}^{-1}$  in the year 2050. Even more pronounced is the increase of the production of organic residues from  $25 \text{ Tg Nyr}^{-1}$  in 1850 to  $125 \text{ Tg Nyr}^{-1}$  in 2000 (Holland et al., 2005). This increase is reflected by an increase of the  $N_r$  deposition on the global scale, even in remote regions (Dentener et al., 2006; Galloway et al., 2008). Since the growth of plants is N-limited in many pristine ecosystems (Vitousek et al., 2002), it has to be assumed that the enhanced N-deposition has caused an increased biomass turnover. It is difficult, however, to determine if and how vegetation-related MA emissions change by such a process. Large-scale land use change has fundamentally altered vegetation patterns (Lepers et al., 2005; Hansen et al., 2010; Klein Goldewijk et al., 2011). Human-induced Climate Change can shift vegetation composition, and climate warming affects plant phenology (IPCC, 2014). It is reasonable to expect that such global changes have altered and will continue to impact vegetation-related amine emissions and their potential contribution to NPF.

Large knowledge gaps concerning vegetation-related emissions of volatile amines exist. The present discussion illustrates the potential of such emissions for a significant contribution to atmospheric amines and makes the link towards their relevance for NPF. Presently, the issue whether the emissions of amines by terrestrial vegetation in-

deed do have an important impact on NPF and associated alteration of global radiative forcing poses an open research question.

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## Appendix: Figure 1: Methods

Twigs with flowering blossoms (*Crataegus laevigata*: approximately 45 blossoms, (*Cornus sanguinea*: 5 umbels) were freshly cut from the plant and enclosed in a small dynamic chamber as used by Kuhn et al. (2011), purged with 0.9 nL lab air with concentrations of approximately  $0 \pm 0.75$  ppb TMA and  $< 7 \pm 1$  ppb  $\text{NH}_3$ , respectively. The outflow was sampled into a HT-CIMS (modified Ionicon PTR-MS in  $\text{O}_2$  mode as used by Kuhn et al., 2011; Sintermann et al., 2011, 2014) for online analysis of  $\text{NH}_3$  and TMA. Calibration factors similar to those used by Sintermann et al. (2014) were applied. Here, the uncertainty range was approximated from an estimated systematic uncertainty of  $\pm 20\%$  in the calibration factors, and considering an additional potential  $m/z$  58– $m/z$  59 interference. Emissions were calculated according to Pape et al. (2009). The given range reflects the estimated minimum and maximum systematic uncertainty. Fractionation of higher-weight compounds to  $m/z$  58, and thus an undiscovered measurement bias, cannot entirely be ruled out. However, we are not aware of such a measurement artefact and generally, the HT-CIMS  $m/z$  58 compared reasonably well to the highly specific results of a PTR-time-of-flight-MS during a measurement campaign in animal housing environments (Sintermann et al., 2014).

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**Table A1.** Literature compilation of MA-enriched species (for another list see Duke and Williams, 1977). Bouchereau et al. (2000) discuss possible measurement uncertainties of amines in plant materials. Some of the older measurements listed in this table might be uncertain due to measurement artefacts.

Species	Investigated	MMA	DMA	TMA	Reference
<i>Anthurus muellerianus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Anthurus muellerianus</i> var. <i>aseroeiformis</i>	fruiting body	x			Steiner (1966)
<i>Arum dioscoridis</i>	condensate in headspace	x	x		Smith and Meeuse (1966)
<i>Arum italicum</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Arum italicum</i>	condensate in headspace	x	x		Smith and Meeuse (1966)
<i>Arum maculatum</i>	spadix	x	x		Klein and Steiner (1928); Stein von Kamienski (1957a); Wink and Hartmann (1981), but contradicting results: see Kite (1995)
<i>Atropa belladonna</i>	leave and blossom tissue	x			Stein von Kamienski (1957a)
<i>Boletus appendiculatus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Boletus edulis</i>	fruiting body	x			Stein von Kamienski (1958); Steiner (1966)
<i>Castanea sativa</i>	blossom odour			x	Porsch (1950)
<i>Chaerophyllum aromaticum</i>	blossom tissue	x	x	x	Stein von Kamienski (1957a)
<i>Chenopodium vulvaria</i>	leave tissue			x	Stein von Kamienski (1957a); Cromwell and Richards (1966)
<i>Chenopodium vulvaria</i>	headspace condensate			x	Wicke (1862)
<i>Ciathrus ruber</i>	fruiting body		x	x	Stein von Kamienski (1958); Steiner (1966)
<i>Claviceps purpurea</i>	sclerotium	x	x	x	Stein von Kamienski (1958)
<i>Conium maculatum</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Corrus sanguinea</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus curvisepala</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus douglasii</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus lucidana</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus insperata</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus jozana</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus laevigata</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus mollis</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus monogyna</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Crataegus spinulosa</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Delphinium consolida</i>	blossom tissue		x		Stein von Kamienski (1957a)
<i>Dermococybe (Cortinarius) cinnamomea</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Dranunculus vulgaris</i>	headspace condensate	x	x	x	Smith and Meeuse (1966)
<i>Geotrichum candidum</i>	arthrospores			x	Maheshwari (2011) and references therein
<i>Gingko bilboa</i>	fruit tissue	x			Stein von Kamienski (1957a)
<i>Heracleum sphondylium</i>	blossom tissue	x	x	x	Stein von Kamienski (1957a)
<i>Hydrosme rivieri</i>	headspace condensate	x		x	Smith and Meeuse (1966)
<i>Iris germanica</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Lactarius deliciosus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Lactarius helvus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Lactarius vellereus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Lepiota clypeolaria</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Lilium candidum</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Lilium martagon</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Lobaria laetevirens</i>	tissue		x	x	Smith (1975) and references therein
<i>Mercurialis annua</i>	leave tissue	x			Stein von Kamienski (1957a)
<i>Mercurialis perennis</i>	leave tissue	x			Stein von Kamienski (1957a); Steiner et al. (1967)
<i>Musa sapientum</i>	fruit tissue	x	x		Smith (1975) and references therein
<i>Mutinus caninus</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Nicotiana longiflora</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Nicotiana paniculata</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Nicotiana rustica</i>	leave and blossom tissue	x			Stein von Kamienski (1957a)
<i>Nicotiana sanderae</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Nicotiana tabacum</i>	leave and blossom tissue	x			Stein von Kamienski (1957a)
<i>Nuphar luteum</i>	blossom tissue	x			Stein von Kamienski (1957a)

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Table A1. Continued.

Species	Investigated	MMA	DMA	TMA	Reference
<i>Phallus impudicus</i>	fruiting body		x	x	Steiner (1966)
<i>Philadelphus lemoinei</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Pholiota mutabilis</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Pirus communis</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Prunus serotina</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Russula alutacea</i>	fruiting body	x	x	x	Stein von Kamienski (1958)
<i>Russula aurata</i>	fruiting body		x		Stein von Kamienski (1958)
<i>Russula azurea</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Russula cyanoxantha</i>	fruiting body		x		Stein von Kamienski (1958)
<i>Russula delicata</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Russula foetens</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Russula grisea</i>	fruiting body		x		Stein von Kamienski (1958)
<i>Russula lepida</i>	fruiting body	x	x	x	Stein von Kamienski (1958)
<i>Russula maculata</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Russula nigricans</i>	fruiting body			x	Stein von Kamienski (1958)
<i>Russula ochroleuca</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Russula olivacea</i>	fruiting body		x		Stein von Kamienski (1958)
<i>Russula pseudodelicata</i>	fruiting body			x	Stein von Kamienski (1958)
<i>Russula sardonia</i>	fruiting body	x	x		Stein von Kamienski (1958)
<i>Russula turci</i>	fruiting body	x	x		Stein von Kamienski (1958)
<i>Russula vesca</i>	fruiting body	x	x		Stein von Kamienski (1958)
<i>Russula violeipes</i>	fruiting body	x		x	Stein von Kamienski (1958)
<i>Russula xerampellina</i> var. <i>barlae</i>	fruiting body			x	Stein von Kamienski (1958)
<i>Russula xerampellina</i> var. <i>rubus</i>	fruiting body			x	Stein von Kamienski (1958)
<i>Sambucus niger</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Sauromatum guttatum</i>	headspace condensate	x	x	x	Smith and Meeuse (1966)
<i>Scleroderma vulgare</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Sorbaria sorbifolia</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Sorbus aucuparia</i>	blossom tissue			x	Stein von Kamienski (1957a)
<i>Splachnum ovatum</i>	tissue	x			Steiner (1966)
<i>Staphylaeae colchica</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Stictia fuliginosa</i>	tissue	x	x	x	Stein von Kamienski (1958)
<i>Stictia limbata</i>	tissue	x	x	x	Smith (1975) and references therein
<i>Stictia sylvatica</i>	tissue	x		x	Stein von Kamienski (1958)
<i>Tetraplodon urceolatus</i>	tissue	x			Steiner (1966)
<i>Thalictrum flavum</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Thea sinensis</i>	shoot tissue	x			Suzuki (1973) (MMA as intermediate in amino acid metabolism)
<i>Tilletia tritici</i>	infested ear			x	Stein von Kamienski (1958)
<i>Trachypus versipellis</i>	fruiting body	x			Stein von Kamienski (1958)
<i>Veratrum nigrum</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Veratrum opulus</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Viburnum opulus</i>	blossom tissue	x			Stein von Kamienski (1957a)
<i>Xerocomus subtomentosus</i>	fruiting body			x	Stein von Kamienski (1958)

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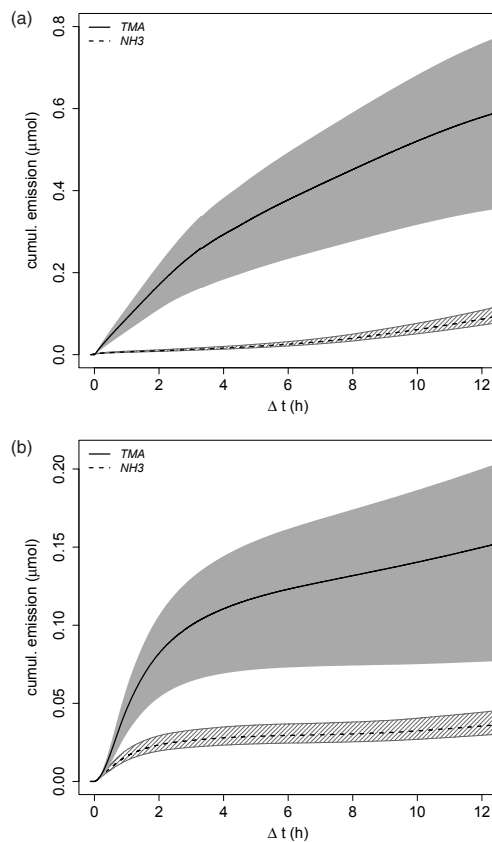
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**Figure 1.** Cumulated TMA and NH<sub>3</sub> emissions from blossoms of (a) *Crataegus laevigata* (approximately 45 blossoms) and (b) *Cornus sanguinea* (5 umbels), determined by dynamic chamber measurements (methods: see Appendix).